Durability measurements of ionic electroactive polymer actuators

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|  | **Abstract** The research is focused on measurements of durability and lifetime of ionic electroactive polymer actuators (IEAP). The aim of the research is developing an universal methodology suitable for comparing the different IEAP types. The principles of the proposed lifetime test methodic and measures of the performance of the actuators are given in conjunction with the evolutionary trends, common to several IEAPs. The tests are carried out using a large scale test equipment, capable to perform the testing process upon many actuators automatically. |

# 1. Motivation

A major goal in soft polymeric electromechanical actuator research is to develop a device of low energy consumption that operates rapidly with high displacement or high force at low applied voltage . The current technologies based on electromechanical and pneumatic actuators make the application products often too noisy, heavy and too complicated. For that reason a notable demand exists for soft, simple and miniature actuation devices in many technological solutions. The ionic electromechanically active polymer actuators (IEAP) are good candidates for this purpose. IEAPs are lightweight and soft multi-functional devices presenting several advantages including the capability of large bending deformation even at low (~1%) intrinsic strains of the electroactive layer and low driving voltages (0.1-5V). In addition to electromechanical bending, an IEAP can sense motion or humidity, harvest energy, and even store electric energy acting as a supercapacitor [1-5]. The possible application areas for IEAPs are the space crafts, medicine, lab-on-chip devices, etc., as this technology can significantly reduce the volume and mass of the equipment. There exists an ample of IEAP-based applications proposed during the recent decade [6-9]. As a rule, all of them have demonstrated their capabilities for a limited time, working close to the performance limit of the IEAPs. However, besides the actual performance of the IEAP devices, there exist more issues, limiting their acceptance in critical applications – reliability and durability.

Reliability has been recognized as the most critical attribute of both critical application areas: space systems and medical systems. Any malfunction of a space mission device commonly means years of work hours wasted and loss of money in the amount of a significant portion of a national budget. Malfunction of prosthesis or a medical device causes serious trouble to the patient. In both cases the worst case scenario may lead to human casualties. Therefore any piece of technology, prior to sending to orbit, or surgically embedding to human body, is carefully tested for the potential causes of failures. Here arises another issue, cutting down the confidence of the reliability tests – the sample size.

It is widely recognized that testing anything, for better confidence the total number of the samples should be large. However, the state of the art in the iEAPs at present time has not reached the mass production or commercial applications yet. Commonly the results published in the scientific papers and reports are obtained upon a few individual samples only. The samples are manufactured in laboratory conditions in small batches, whereas the fabrication resembles cooking, requiring know-how, experience, and skillful hands. Only a few researchers dare to admit honestly that the production is irregular [10], while it can be argued that no two samples are exactly alike. As a result, the situation of the IEAP industry is very different from that, say, of the confectionery industry, where millions of identical biscuits are always available for testing. Nevertheless, in our lab, in the course of the process of fine tuning the IEAP fabrication recipes, the pile of IEAP samples of various kinds grew up to the quantities difficult to handle. In order to compare the individual samples and perform the lifetime tests we developed a large scale test equipment to perform the testing process upon many actuators automatically. This setup excludes the human errors and guarantees the all samples are tested in exactly similar conditions. Moreover, this setup is able comparing different IEAP types, carrying out the measurements using identical methodology.

During the last two decades the assortment of IEAPs has evolved from the aqueous ionic polymer-metal composite (IPMC) and “wet” conducting polymer actuators to a rich choice of various types. There are published thousands of papers about the various IEAPs in the recent decade reporting about the better strain and speed, or smaller dimensions. However, only a few of them tell about the lifetime or cycle life of their samples, while the reported data is beyond compare. It is commonly recognized that lifetime, or cycle life of IEAPs is highly strain- and stress dependent, while frequency dependence can arise due to dissipation and heating [11]. At large, the reported cycle life of different IEAPs varies between thousands and millions of cycles. Some authors praise the large strain of their production, but the cycle life tests are implemented at high frequency and almost imperceptible strain. Obviously, the reason of the data discrepancy is the absence of a generally recognized methodology.

From the viewpoint of the cycle life, the most thoroughly studied IEAPs are the conducting polymer actuators. Vidal et al. inform that their actuators standing of interpenetrating polymer network actuators containing an electronic conducting polymer can be cycled for 104 times at 1 Hz, and 3.5×106 times at 10 Hz [12]. Lu et al. just stop the lifetime test of the π-Conjugated Polymer devices after 104 cycles [13]. Madden et al have performed thorough research and argue that the cycle life of the polypyrrole actuators depends on the strain, reaching 103 cycles at high strain and 32×103 at low strain at the frequencies of 0.2-1 Hz [14], and 1.2×105 cycles with very low strain at 3 Hz [15]. Liu et al describe the loss of performance of similar actuators as 20% loss after 104 cycles, 50% at 3.5×104, and virtually no electroactivity by 6×104 cycles [10]. On the contrary, Bennett and Leo compare the traditional nafion-platinum IPMC with water and ionic liquid as electrolyte, and report the cycle life being 103 for the former and 3×105 for the latter [11]. According to Du et al. their ionic polymer–carbon nanotube composite actuators can last for 3000 cycles with only 10% reduction in the displacement output [16].

The purpose of the paper at hand is sharing the experiences and best practices emerged in the course of designing and use of the automatic equipment of testing IEAPs. As its design and capabilities are based on the trade-off between the automation level, ease of use, and the acquired data, the focus of the current paper is justifying the design principles. In terms of the long-term degradation of the IEAPs, the evolutionary trends, common to all IEAPs, rather than the comparative data analysis or formal statistics of all particular samples, are given. The described testing methodology describes well degradation of IEAPs, hence we recommend it as the standard procedure.

# 2. Method

## 2.1. Pre-requisites

In general, the performance of the IEAPs decreases in time. The rate of performance drop depends on a variety of processes such as frequency, number of performed work cycles, applied load, strain, stress, but also on the environmental conditions: temperature, humidity, etc. It is likely that some of the factors remain yet unknown.

The long-term experiments with IEAPs show two types of degradation:

* degradation during operation – the fatique of the membrane and electrodes as well as leakage of the electrolyte out of the membrane caused by its mechanical deformation;
* spontaneous self-degradation – long-term alteration of the parameters, caused by the ambientenvironment. We noticed the spontaneous self-degradation even when the samples were held in the ideal conditions suggested by the manufacturer, e.g. immersed in the electrolyte.

## 2.2. General test procedure.

In order to detect the presence of the two types of degradation, the long-term test was carried out in the following order:

1. Testing the initial condition of each particular sample;
2. In 10 weeks the condition of each particular sample was recorded again;
3. In 10 weeks from the Step 2 the lifetime of the samples under continuous loading was determined. This process was terminated when the performance of most of the samples was below some pre-determined value, commonly 5% of the mean value of the initial values.
4. In about 2 weeks after completing the Step 3 the final condition of the samples was recorded once again.

Between the steps the samples were kept in ideal environmental conditions – each one in a separate vial, in the appropriate solvent, if required.

The lifetime measurements were carried out using identical methodology upon the different IEAP types. The shape of the exciting signal, timing, and even the dimensions of all samples were identical – 5 x 20 mm. The only parameter that was dependent on the type of actuators was the amplitude of the excitation voltage, determined by the electrochemical window of the particular IEAP type.

As the expected lifetime, sometimes referred to as “cycle life”, of the actuators is 104 ... 107 working cycles, it is not necessary to measure just each cycle. It is enough to test each sample after a while of working. Therefore each sample is swapped between the measuring station and training station. During training, the actuator is loaded with the appropriate voltage input but its electrical and mechanical responses are not recorded. During measurement the actuator is engaged with signal of wide frequency range, while the mechanical and electrical parameters of interest are recorded. The relatively high cost of the components (camera, displacement sensor, force gauge, current booster, data acquisition system, etc.) of a measurement station justifies dividing the equipment to expensive test station and inexpensive training station. On the other hand, swapping between the two stations makes the whole procedure quite labor-intensive. Testing a single actuator manually consists of the following steps:

1. The actuator is attached to the electric contacts. The samples are somewhat fragile, therefore should be handled carefully. If the sample is reattached to the contacts several times, the operator must ensure that every sample is always oriented similarly and that the length of the sample between the contacts must be always similar.
2. The camera or laser displacement sensor, recording the bending response of the actuator, is focused to the sample. The actuator is excited with the pre-defined test signal. Simultaneously, the mechanical output, input voltage and input current of the actuator are recorded.
3. The camera is replaced by a force sensor. The blocking force is measured only in the straight position. Analogously to the previous, the actuator is again excited with the similar test signal, and output signals are recorded.
4. The actuator is detached from the measurements station, and attached to the separate training station, where all actuators are excited with similar sine voltage of constant frequency. Special care is paid to the accounting of the exact number of passed working cycles of each particular sample.

This process is continuously repeated until decided to terminate.

# 3. Large Scale Test Equipment

## 3.1. Working principle

The described testing procedure is a tough and precise work for an operator. One operator is able testing concurrently 10-20 samples only, performing a few tens of cycles per day. Therefore we designed an authentic equipment to perform this process upon many actuators automatically. This setup excludes the human errors and guarantees the all samples are tested in exactly similar conditions. Its design and capabilities are based on the trade-off between the automatization level, ease of use, and the amount and type of the acquired data.

The photos of the test bench are given in figures 1 and 2. It is a circular conveyor of 4 storeys, sharing the same rotary axis and acting synchronously. Each storey can hold up to 60 actuators of similar type, so the whole bench can hold up to 240 samples of up to four different types. The actuators are attached to the clamps, hanging around the turntable during the whole measurement process. This method avoids their damage and ensures that the samples are not turned over between the cycles.

The equipment was located in a separate room equipped with an air conditioner of sufficient capacity to guarantee the stable temperature (17 ºC) and humidity level (30 % RH) of the ambient air for the duration of the whole experiment.

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| Figure 1. The 4-storey conveyor. | Figure 2. Two storeys of the test bench with hanging clamps. |

Concurrently 4 samples are measured by the cameras (one for each storey); and 4 samples are measured with the force sensors (one for each storey). After performing the measurement cycle, the circular conveyor turns for 6 degrees, sets the 8 samples, just tested, for training, and positions the next 8 samples for testing.

Measurements are carried out in cantilever configuration - one end of an actuator is fixed while the rest is free to move or to apply force to the force gauge. During the measurement cycle the sample is held horizontally and edgewise, in this configuration the mass of the actuator itself distorts less its free bending. During the training cycle the sample is held hanging vertically. This is done for two purposes. Some IEAP types exhibit considerable creep - permanent deformation under the influence of the inner stresses. When the actuator is hanging freely, its own weight diminishes this effect. Furthermore, some types of IEAPs must be continuously in the wet environment, others have to be moistened in water or some solvent after every certain time. Therefore the vertical placement of the samples allows soaking the wet types of IEAPs in their solvent bath. To avoid drying of the “wet” actuators, the measurement cycle that is carried out in ambient environment should not exceed a few minutes, covering all required frequencies. An example of an actuator, lifted out of the wet environment, is depicted in figure 3.

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| Figure 3. For measurement a sample is lifted out of the water bath. The proper illumination is essential. |

3.2. Technical details

Only the samples under testing are positioned horizontally while all remaining samples under training are hanging vertically. The process of turning the samples between the two positions is explained in figure 4. Initially the sample is hanging vertically, possibly in a container of solvent, while the turntable squeezes the electrical terminals of the clamp against the properly positioned electric contacts (figure 4A). In order to align the sample horizontally, the turntable with all clamps lifts up and turns, positioning the particular clamp over a tubular support, named “lower support”. The lowering turntable forces turns the clamp up (figure 4B), until the electrical terminals are squeezed against the appropriate spring contacts (figure 4C). After that, the sample is ready for testing - in the cameras field of view or pushed against a force sensor. The state of the remaining 58 samples is as in figure 4A. Aligning the sample back vertically is performed by lifting the turntable up, and pushing the clamp against the upper tubular support. Next, the turntable is rotated by 6 degrees, while lifting it down arranges the just measured sample in the alignment depicted in figure 4A, and sets the next one under measurement (figure 4C). This mechanism allows switching between two alignments synchronously 8 samples – two on each of the four storeys. This way, rotation of the turntable enables measurement of all samples, one after another. The videos showing repositiong of the samples are given in the online supplementary materials.

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| 1. Training cycle. Actuator is positioned vertically | 1. Lifting sample up. |
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| 1. Measurement cycle. Actuator is positioned edgewise and horizontally. | 1. Lifting sample down. |

Figure 4. Repositioning the samples between vertical and horizontal alignments.

Special attention was paid to the contact clamps. In order to avoid contamination of the electroactive materials, the contacts should be made of a noble metal – gold or platinum. Initially the clamps were fabricated using the technology of printed circuit boards. The gilded electrode surfaces were obtained by standard surface plating used for printed circuit boards – electroless nickel immersion gold (ENIG) method. However, according to the manufacturer’s data, the thickness of the layer of gold, plated using the ENIG method, is 0.05 – 0.125µm only, while under it is a 3-6 µm nickel layer on a 35 µm copper. As shown in figure 5A, even a gentle touch can rub off the thin gold and reveal the electrochemically active nickel or even copper. For that reason the contact clamps were reworked, soldering to clamps a 1.5 mm wide ribbon of 50 µm thick sheet of gold. The solder and whole opposite surface was covered by a resistant epoxy lacquer. The contact clamps of the “best practice” are depicted in figure 5B.

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| A | B |
| Figure 5. Contact clamps. A: unsuitable; B: best practice. The width of the contact clamp is 8 mm. | |

## 3.3. Recorded mechanical output

The recorded mechanical outputs were the output of the force gauge and the information of bending. The most reliable way of describing the behavior of IEAPs is processing the visual information of bending. The bending behavior of the actuators was recorded by cameras, particularly the monochrome USB cameras DMK 22BUC03 equipped with a lens of long focal length to reduce the effect of parallax. The proper illumination of the samples from back and from two sides as depicted in figure 3 simplifies the latter image processing.

For force gauges, the Millinewton force sensors of EPFL, and MLT0202 of ADInstruments were used. Measuring the force may seem a simple and cheap task, as the output of the force sensor is easily registrable voltage. However, automated positioning of an actuator to a force gauge is a complicated procedure as a clumsy manipulation may easily damage the samples, especially the samples suffering from the creep. The illustrative examples of such warped actuators, given in figure 6, are fully viable, despite of their initial twisted shape. For such types of actuators the force gauge was removed in order to prevent their damage.

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| Figure 6. Actuators suffering from creep, in their initial unengaged state. |

## 3.4. Schematic of the measurement

The schematic of the experiment set-up is given in figure 7. Similar configurations are described in our previous works [17, 18]. In order to reduce the load of the computer, the voltages are generated by specialized generator using an Atmel microprocessor. The generator produces the AC signal for driving the actuators, and the trigger for synchronizing camera and he DAQ device. Using this special generator allows synchronizing the measurements and camera with the input signal in the minimal sampling rate for each particular frequency. Thus the number of frames captured by the camera and the number of acquired samples in about 2.5 minutes of measurement were reduced to 170 and 700 respectively. The input signals are amplified by custom made current amplifiers, based on the Burr-Brown OPA548 op amp. Altogether, the 4-storey conveyor incorporates:

* 5 special generators: one for each storey, and one for generating the common training cycles;
* 8 current amplifiers, two at each storey: one for measurements and one for training;
* 4 USB cameras: one for each storey;
* 4 force gauges: one for each storey.
* 1 motorized linear actuator Thorlabs TravelMax Stage LNR50VK1/M to lift the circular turntable up and down within 5 cm;
* 1 motorized rotational stage Thorlabs CR1/M-Z7E to rotate the turntables continously with respect to the vertical axis.

All electrical signals – voltages, electric currents, and outputs of the force gauges are registered by a single National Instruments NI PCI-6254, M Series DAQ card with 32 analog inputs. The electric current is measured as a voltage drop over a low-ohm resistor. The whole system is operated by a single PC-type computer, and the National Instruments LabView software.

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| Figure 7. schematics of the 4-storey conveyor. |

## 3.5. Driving signal

Measuring the „wet“ IEAPs in dry environment should be performed as fast as possible, while in the case of „dry“ EAPs there is no such restriction. For example, the conventional aqueous ionic polymer-metal composite (IPMC) can be in air up to a few minutes before its parameters change drastically due to drying. For our synchronous 4-storey conveyor, the „wet“ IEAPs require that the cycle of measurement should not exceed 2-3 minutes. To cover wide frequency range, the driving voltage signal was a gradual sweep lasting 2.5 minutes consisting of series sequence of sine signals of different frequencies. Each frequency lasts 3 half periods. The delay between the sequences was at least 0.5 half periods of the previous frequency while the initial phases were opposite. The frequencies were (in this order) 10, 5, 2.5, 1.25, 0.64, 0.32, 0.16, 0.08 and 0.04 Hz.

## 3.6. Reducing the amount of acquired data

The set of the data acquired during the measurement of a single sample involves the analog signals such as voltage applied to the actuator, consumed electric current, and the output of the force gauge. As an example, typical set of acquired signals of a force measurement is given in figure 4A. We see that the signal of the force gauge is asymmetrical. This is caused by improper automatic positioning of a twisted sample.

In order to describe behavior of the actuators in time, we needed a parameter, describing their performance. A comprehensive overview about the commonly used quantitative representations is given in [19]. As the total number of IEAP samples is quite large, an important requirement fo this parameter is the possibility to perform the measurements automatically, regardless of the behavior of each sample or each particular IEAP type. For estimating the performance of the bending IEAPs actuators, the commonly used parameters are the strain difference and tip displacement. Both of them can be used in the case of small deflections only. In order to describe large bending of the actuators, we characterize the shape of the actuators by a set of vectors. Similar methodic is described in our previous works [17, 18].

The vectorial interpretation of the shape of the actuator expresses its bending with respect to the distance from the input contacts along the sample. The curved line representing the shape of the actuator is divided into vectors of equal lengths as depicted in figure 8, assuming that within every vector the curvature is constant. The shape of the actuator is characterized by a number of angles θ1, θ2, … θn. Each consecutive angle along the length of the actuator is relative to the previous one. The length and total number of the vectors is a trade-off between the accuracy of the result and the resolution of the camera. It is self-evident that more vectors represent a complicated shape better. However, the relative error of detecting the angle depends on the resolution of the recorded image In our experiment the resolution of captured images was 640×480 pixels, while the actuators were divided into 9 vectors.

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| Figure 8. Shape of the actuator is described by angles **θ1, θ2, … θn** and performance **β**. |

The image of the actuator is recorded by a USB camera (particularly the DMK 22BUC03 was used), and processed by the appropriate National Instruments LabView tools. A proper lighting and exposure as shown in figure 3, greatly simplifies the procedure of determining the vectors. When the direction of the camera is set transverse to the actuator and the stage is illuminated from the background, the image of the actuator becomes a single contrast curved line. The side illumination, as depicted in figure 3, further eliminates even the shadows emerging due to the actuator twisting.

An experiment setup like described above, generates enormous amount of data, if data acquisition rate for the whole procedure is chosen based on the highest frequency. The required disk space, especially for storing the video information, easily grows into terabytes. Another critical issue besides the storage challenges, is the bandwidth of data bus given that 4 USB cameras were used. In order to reduce the load on the main PC, stand-alone signal generators were designed and implemented.

These signal generators also produce the trigger signal to synchronize the camera and the driving voltage. This setup allows capturing video with a dynamic frame rate that adapts to the particular frequency of input signal. The number of frames captured by the camera during the approximately 2.5 minutes of measurement was, thus, reduced to 175. However, as the file size of such a video was around 50 MB, the images were converted to the vector representation immediately after the measurement cycles. Discarding the images helped to reduce the amount of required storage space significantly as the data describing a single measurement was condensed to a few dozens of kilobytes.

While the obtained set of vectors allows exact reconstruction of the shape of the actuator during the measurements, it is still too large to effectively describe the degradation level of the actuator. Analysis of the obtained data showed that a convenient quantitative parameter to evaluate the performance the condition of an actuator is the angle between the tip tangents in the case of maximal bending displacements to the opposite directions. It describes well the behavior of an actuator of given length and as all the tested IEAP samples were of same length, adequate comparison of performance for all the actuators is possible. As depicted in figure 8, this parameter, named as β , will not fail even in the case of considerable creep of the sample. As example, the typical sets of acquired data during the force and video measurements are given in figures 9 A and B respectively. The figure 9A shows the typical shortage of the force measurement – force, generated by an initially curved actuator is detectable only partially. For that reason, hereinafter we rely on the video data only.

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| A | B |
| Figure 9. A – Force measurement; B – video measurement. For clarity, all signals except voltage are scaled | |

# 4. Long-term measurements

In the scope of current paper we present and analyze long-term degradation of two different types of IEAP fabricated onsite, with carbonaceous electrodes and with conducting polymer electrodes:

Acronyms and abbreviations used in this chapter:

* CDC – nanoporous carbon, derived from titanium carbide;
* EMIBF4 – 1-Ethyl-3-methylimidazolium tetrafluoroborate;
* LiTFSI – lithium bis(trifluoromethane)-sulfonimide;
* MEG – monoethylene glycol;
* NaDBS – sodium dodecylbenzenesulfonate;
* Na2S2O8 – Sodium peroxodisulfate;
* PC – propylene carbonate (4-methyl-1,3-dioxolan-2-one);
* PVdF – poly-1,1-difluoroethene, polyvinylidene fluoride;
* PVdF(HFP) – poly(vinylidene fluoride-co-hexafluoropropylene);
* Py – pyrrole;
* Ppy – polypyrrole.

1. A PVdF(HFP) membrane with electrodes consisting of CDC. The electrolyte is EMIBF4 ionic liquid.

The electrolyte of this IEAP is a nonvolatile ionic liquid, while the main component of the electrodes is extremely absorbent porous carbon. Therefore it is intended to work only in dry environment without adding the electrolyte. The amplitude of the driving voltage was the recommended ultimate working voltage - 2.8 V.

Preparation: the electrode layer, composed of CDC powder, EMIBF4, PVdF(HFP) was made by casting the suspension into a Teflon mold and then drying at 80° C in a vacuum oven. The electrolyte film was obtained by pouring the mixture of PVdF(HFP) and EMIBF4 into the Teflon mold and evaporating the solvent completely. Finally, the electrolyte film was sandwiched between two electrode films and hot-pressed to connect the layers.

For the details and fabrication, see e.g. [20].

1. A PVdF membrane with polypyrrole electrodes and with PC+LiTFSI (1.0M) electrolyte, fabricated by the combined chemical and electrochemical synthesis method [21].

This IEAP is capable working in air, but in order to avoid drying, it is recommended to soak it periodically in the electrolyte solution. In the course of the long experiment, the delay between moistening procedures was 110 minutes. The amplitude of the driving voltage was 2.0V.

Preparation: commercial Millipore PVdF membrane (according to specification: hydrophobic, thickness 125 µm, pore size 0.45 µm, porosity 70%) was used as electrode storage layer. Electrodes for electrochemical synthesis were synthesized chemically as follows: membrane was permeated with pyrrole monomer and immersed in 0.006 M NaDBS, 0.075 M Na2S2O8 aqueous solution at 60 °C for 45 s; the polymerizing Py from outer-most pores turned the membrane black. The chemical synthesis was terminated by washing membrane with cold methanol. PPy was deposited galvanostatically on both sides of the membrane. Electrochemical synthesis was carried out galvanostatically in an one-compartment two-electrode electrochemical cell. The synthesis solution contained 0.2 M Py and 0.2 M NaDBS dissolved in a mix of water and MEG.

## 4.1. Electromechanical impedances

In the course of the long-term experiment, measurements resulting with graphs as depicted in figure 9, were taken after every 174 training cycles. The obtained data allows describing the performance of the actuator until its degradation at each particular frequency. The 3 impedances – voltage vs. electric current, performance vs. voltage, and performance vs. current - of arbitrary samples of IEAPs of both IEAP types during their whole life, are given in figure 10.

In figure 10 we see that the in 8000 working cycles the electrical impedance voltage/current, or the internal resistance of the actuator, increases about one magnitude. Similar one-magnitude decrease shows the relation tip angle/voltage. However, the relation between the tip angle and electric current remains constant in the whole frequency range. Analyzing the data we have found that observing the electric current gives adequate information about the degradation level of all samples investigated. Moreover, as we show hereinafter, this electrically measurable parameter can indicate the sudden failure in the case of conductivity break-off as well as short-circuit of an actuator.

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| Type A, voltage vs. current | Type B, voltage vs. current |
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| Type A, performance vs. voltage | Type B, performance vs. voltage |
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| Type A, performance vs. current | Type B, performance vs. current |
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Figure 10. 3 Impedances of the IEAPs of two types in the course of degradation.

## 4.2. Long-term degradation of the IEAPs.

The electromechanical impedances presented in figure 10 show that the tip angles β evolve in a similar manner in the whole frequency range. For that reason it is justified to only look at the long-term performance of a single - in our case the lowest – frequency, brought forward in the figure 9B.

The obtained performance data can be plotted in respect to time as well as to performed work-cycles for each particular sample. The former emphasizes the spontaneous self-degradation while the latter shows degradation during operation.

figure 11 presents the plots of two samples of the IEAP of type A with respect of the two coordinate axes. One of them suffers from an occasional damage between the days 100-120. The time-dependent behavior shows that the performance of the actuators decreases even in the idle state. The Step 4 (see the section of the general test procedure hereinabove) shows that degradation continues with the similar rate after the long experiment is terminated. The graph of the performance versus number of performed working cycles shows that performance decreases nearly exponentially. In spite of many efforts we could not find any functional fit of degradation, matching all samples or all IEAP types with sufficient accuracy.

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| Figure 11. Pair of degradation charts of two arbitrary samples of the IEAP of type A. Each point in the graphs marks one measurement cycle. To each point in the left graph corresponds one point in the right graph and vice versa. | |

## 4.3. Occasional damage

Although the objective of the current project was determining the durability of the IEAPs, this experiment of large scale gave several unexpected results. Here we present some of them.

* Besides the progressive weakening of the performance, some occasional samples exhibited sudden destruction, independent on the environmental conditions. This behavior was observed in the case of all IEAP types. The corresponding degradation graphs with respect to the number of cycles passed are presented in the figure 12 A and B. Tracking the corresponding behavior of electric current and the electromechanical impedance shows that there exist two ways of sudden destruction:
  + Conductivity break-off (figure 12A). The sharp decrease of the performance is accompanied by sharp decrease of the electric current. This is caused mainly due to the delamination of the electrodes. The scanning electron microscope (SEM) micrograph of a delaminated part of the IEAP A is depicted in figure 13.
  + Short-circuit of the sample (figure 12B). The sharp decrease of the performance is accompanied by sharp increase of the electric current. The close inspection showed that the short-circuit happens exclusively due to the permeation of the overheated membrane and the electrolyte between the electric contacts. The SEM micrograph of a melt part of the IEAP of type A is presented in figure 14.

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| A | B |
| Figure 12. Two ways of the occasional damage of IEAPs. A: conductivity break-off after 4200 cycles; B: short-circuit after 5900 work-cycles. Each point in the graphs marks one measurement cycle. | |

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| Figure 13. Delaminated electrodes of the IEAP of type A. | Figure 14. Partially melt membrane of the IEAP of type A. |

Analyzing the data we found that some IEAP samples regained their performance at long time after sudden destruction. The corresponding pair of degradation charts is given in figure 15. Normally, when experimenting with IEAPs, the destructed samples are considered being unsuitable for further experiments. In our long-term degradation experiment none of the samples were removed from the conveyor before total termination of the experiment. Analyzing the data we have observed that some samples have regained their performance after several thousand ineffectual excitations. The distinctive behavior of electric current gives reason to believe that the melt electrolyte, stained near the electric contacts, finally dries out, and eliminates the short-circuit.

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| Figure 15. Pair of degradation charts. This sample suffers from short-circuit after 320 cycles, revives after 5000 ineffectual tries, finally after 2000 effectual working cycles breaks down again. . Each point in the graphs marks one measurement cycle. To each point in the left graph corresponds one point in the right graph and vice versa. | |

## 4.4. Rate of use

We found that the lifetime of the IEAPs depends on many processes, terms and conditions: the amplitude, frequency, and signal shape of the applied signal, temperature and humidity of the ambient environment, etc. Moreover, it depends even on the rate of use. To demonstrate this phenomenon we performed an experiment with two identical samples of the IEAP of type B (with polypyrrole electrodes). Actually, to obtain a pair of identical samples, a 5x20 mm sample was cut into halves lengthwise. One of them was subjected to a continuous repetitive test cycle, while for the other the delay between the test cycles was one hour. The resulting degradation charts given in figure 16 show that the lifetimes of the samples are different by means of the performed working cycles as well as by means of time passed. The performance of the continuously working actuator falls to 0.05 in 8 hours after performing 3300 working cycles. The less frequently actuated sample weakens to the same level after performing only 400 working cycles in 50 hours.

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| A | B |
| Figure 16. Degradation of a sample of the IEAP of type B at different utilization rates. A: continuously; B: in hourly delays. | |

# Discussion

We have developed a methodology for testing durability of IEAPs, taking into account the two types of degradation - degradation during operation and spontaneous self-degradation. The experiments carried out by this methodology and the large scale equipment show that both types of degradation have serious effect to the IEAPs. Here comes the justification of the term “lifetime” of IEAPs over the term “cycle life”. In our opinion the described methodology is a good candidate to become a standard procedure of durability testing.

Though we have performed the long-term degradation experiments on many types of IEAPs, in the current paper we limit the discussion about the results with a few examples only. The reason is absence of a correct methodology of the failure distributions characterization. We must recognize that the performance loss and sudden destruction of IEAPs is rather random. In spite of many efforts we could not find any functional fit of degradation, matching all samples or both IEAP types with sufficient accuracy. Similarly, the distribution of lifetimes cannot be well described by any of the commonly use methodologies of failure distributions characterization, e.g. the Weibull statistics. This deduction is in full agreement with Liu et al. [10].

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