**Novel model of viscoelasticity for IEAP actuators.**

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**Abstract** tuleb alles siis kui conclusion on valmis.

The innovatory viscoelastic model of bending ionic electroactive polymer actuators allows modelling

**1. Introduction**

Already since the very first reports concerning the bending ionic electroactive polymer (IEAP) actuators, researchers have described the back-relaxation effect – an actuator excited with DC voltage, instead of holding its bent state, relaxes slowly back towards its initial shape. This behavior is commonly treated as a shortcoming of IEAP actuators, decreasing their ability to deliver a constant peak force to target, limiting their frequency range, and hindering their exact control. There have been several attempts to slower the rate of back-relaxation by choosing an opportune combination of separator, electrodes, or mobile ions. Nevertheless, there is no report claiming that, in the case of some IEAP material, this holdback is completely eliminated.

Supposedly, the phenomenon of back-relaxation is studied almost only in the case of „old-type“ ionic polymer-metal composites (IPMC). It is a wet ion exchange polymer membrane covered with thin metal electrodes on both sides. [X7] For typical IPMC with platinum electrodes, when a voltage is applied across the thickness direction, the electromechanical transduction occurs due to the motion of the cations along with the water molecules. As a result, one face of IPMC withers and shrinks, while the opposite face protracts due to the swelling of water and ions. This generates warping of the whole laminate. The resulting fast bending motion toward the cathode is followed by a slow relaxation towards the anode, while IPMC as the whole still remains bent. If now the input of this actuator is shorted, the actuator performs a sharp movement towards its initial state, but will usually outdistance. The steps of this process are depicted in figure 1. The speed of back-relaxation is reported being dependent on the type of polymer membrane, solvent, cations, etc. [X6]. It is commonly believed that the slow relaxation happens due to the water diffusion out of the strained polymer matrix. [X7] Most of the proposed electromechanical models describing the behavior of IPMC, just neglect the phenomenon of back-relaxation. [XXX] One of the few exceptions is presented by Bao et al. [X3], proposing the model of IPMC without and with the back-relaxation by adding a separate relaxation time constant. [X8] kirjeldab ka midagi.

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| (a) | (b) | (c) | (d) |

**Figure 1**. Actuation of IPMC. (a) – initial relaxed state; (b) – forward bending response to a constant input voltage; (c) – back-relaxation with voltage still applied; (d)-opposite bending due to shorted input. The watch serves for time monitoring.

In recent years the range of IEAP materials has been significantly extended by means of materials used for electrodes, separator membrane, as well as the electrolyte. At first glance, all ionic EAPs seem similar in construction – two conducting electrodes separated by a polymer membrane, containing freely moving ions – but their actuation mechanisms can be significantly different. There haven’t appeared any definitive models or at least reports yet, describing the phenomenon of back-relaxation of IEAP actuators with electrodes based on carbon, or conductive polymers.

Experimenting with the IEAP actuators with carbon electrodes, we have noticed that similarly to the water-containing IPMC, virtually all of them exhibit back-relaxation, at least to some extent. It appears that by means of their transient spatial actuation and transient moment of force, the long-term behavior of all IEAP actuators is just similar. The back-relaxation is present regardless of the absence of the fluent liquid, and regardless of the membrane or electrode material.

The bending-relaxing behavior of IEAP actuators resembles viscoelasticity, but seems working partly conforming, partly contra to the electrical excitation. For this behavior, the constitutive models of viscoelasticity, consisting of linear combinations of springs and dampers, are inappropriate. There exist numerous models describing the material’s response in different loading conditions, e.g. Maxwell model, Kelvin-Voigt model, or combinations of both of them. [XX] All the models describe only the response of the material to the external load. Alas, neither of them involves any element causing the actual behavior of the IEAP actuators – lapsing against the load. The cause of this failure is the concept of the so-called canonic models of viscoelasticity [XX], where the load is external, and is applied to the whole system.

The objective of the current paper is to describe the time-dependent correspondence between the transient input voltage and the shape of the IEAP actuators, taking into account the back-relaxation. We ground to the scalable distributed model of IPMC, presented in [DM], and extend it to different IEAP materials. This approach describes the commonly uneven propagation of voltage along the IEAP material, and associates the voltage and charge carried over with the uneven flexure of the actuator. The experiments described in [DM] assume that the charge-flexure relation is simply linear, and leave off just before the phenomenon of back-relaxation becomes noticeable. The current paper associates our novel model of viscoelasticity with [DM]. The resulting electromechanical model of IEAP allows describing the shape or blocking force of the actuators over a long time. Its transient input signal is not limited in any way, due to the accordance to the Boltzmann superposition principle – each loading step makes independent contribution to the total loading history [ISBN: 1569903484].

This paper is organized as follows: Firstly, in Section 2 we show that the innovatory approach to an assembly of springs and dampers will result with PDE-s describing the truthful bending of the actuators forwards and backwards under a constant exciter. In Section 3 we demonstrate the capabilities of the proposed model of viscoelasticity with the response of a lumped model of a fictional IPMC. The thorough characterization of the mechanics of IEAP actuators given in Section 4 is inevitable for understanding the distributed electromechanical model introduced in Section 5. The verification of the model along with the examples of three completely different IEAP materials is presented in Section 6. The discussion in Section 7 disputes about the further improvements of this representation.

**2. The model of viscoelasticity with internal strain.**

As mentioned hereinabove, in the canonic models of viscoelasticity the load is external, and is applied to the whole combination of springs and dampers. This configuration does not involve even any possibility to suppress the exciter. The situation changes completely, when the exciting factor is applied between the spring and damper, resembling some factor expanding-contracting between the elastic threads of the polymer molecules network. The viscoelastic scheme is depicted in figure 2. It consists of a damper in series with our new element - internal strain -, both in parallel with a lone spring. The internal strain is a vector of variation of the length of a non-elastic element. Hereinafter we associate it with the transient electrical charge **q** of the IEAP material, therefore the strain element is labeled as **Q** and portray as **Q** between arrows for clarity. When internal strain of the element **Q** is applied, the spring reacts momently, while the damper follows with time, tending towards the unstressed state of the spring. In brief, the input of our viscoelastic element is the strain , while the output is the strain of the spring .



**Figure 2**. The model of viscoelasticity with internal strain.

The behavior of this viscoelastic model in different loading conditions is depicted in Fig. 3. We insist that there are no other external stresses or strains affecting the system besides, indicating that. Initially, the spring and the damper are in their unstressed states: (a). The positive strain of Q draws the spring out (b) while the damper releases in time. This process continues regardless of the applied strain Q until the stress of spring is vanished (C). The removal of the strain Q results with the compression of the spring (D), followed by gradual dragging of the damper, until the stress of the spring is again zero (E). The negative strain of Q is depicted as its shortening (F) and compression of the spring. Similarly, the spring draws the damper longer until reaching the unstressed state (G). As seen in figure 3, the assembled system strives to gain its unstressed state with any stress Q.

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| (a) | (b) | (c) | (d) | (e) | (f) | (g) |

**Figure 3**. Response of the model in different loading conditions.

The posed model is defined with four relations in (1).

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where and are modulus of elasticity of the spring and viscosity of the damper respectively. We see two equivalent solutions to this model: with respect to the input strain and with respect to its derivative, here we present both of them. The described relations in (1) can be arranged into differential equation for strain of damper.

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The solution for is found by bearing in mind and solving the first order linear differential equation for with initial condition

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A different form of the equation is derived by combining (1) for strain of the spring directly

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Solving (4) with gives

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This form might be useful if the derivative of is known. By introducing the rate of relaxation and expressing the input strain in terms of stress both given forms are simplified to (6) and (7).

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**3. Lumped model of IEAP with relaxation**.

In order to demonstrate the capabilities of the proposed model of viscoelasticity and its correspondence to the superposition principle, we use a simple lumped model of IPMC presented e.g. in [Bao]. That concept couples the input voltage with transferred charge and the tip displacement of an IPMC actuator. It gives reasonable results in case of small displacements only but serves well to explain the complicated approach presented in the next sections of this paper. Here we just demonstrate the transient response of the model to a rectangular input signal without attempting to validate it with any real actuator.

The lumped model of IPMC is presented in Fig. 4A while the electrical signals as well as strains are depicted in Fig. 4B. When voltage Uin is switched on, the charge curve of the capacitance is exponential with respect to time, and the correspondence between the strain (or tip displacement) and charge is proportional: ε(t)=k\*q(t) . The diagram Fig. 4B depicts the rectangular voltage Uin, the charge of the capacitance q and two according to (6) numerically modeled transient strain behaviors with different parameters λ and convenient values of the coefficient k to fit the graphs into the picture.

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| (a) | (b) |
| **Figure 4**. Scheme and behaviour of lumped model of an IPMC. | |

Supposing that initially the system has been shorted for long time, the initial charge of is zero: . When voltage is turned on, the strain curve initially follows the charge curve , turns apart when the integral part of (6) grows, then relaxes slowly with the speed determined by . When the voltage is switched off, the actuation towards the initial state again keeps the curve of charge followed by back-relaxation, but according to the superposition principle starts from the momentary values of strain and charge. Similar back-relaxing behavior of IPMC is described in numerous papers e.g. in [Bao, XX1,XX2,XX3].

**4. Distributed bending moment of the actuator**

Describing the bending IEAP actuators, it is convenient to observe only its short segment, for instance its tip. So far, lots of the publications follow this pattern and model the tip displacement in Cartesian coordinates, the blocking force of the tip, etc. [XX,XX]. These approaches assume that the shape of the actuator is characterized with a single radius of curvature, and the strain of the actuator reflects directly the tip displacement. This unsophisticated concept can be true in the case of small unobstructed displacements only. As soon as the movement of the actuator is restricted, for instance by the gauge measuring the blocking force, its shape will deviate from circular, and the whole concept fails. The vectorial interpretation presented in this section is universal and serves as the first step in modeling the distributed bending moment of the actuators independently of their actual shape.

**4.1. Characterization of the shape of the bending actuator**

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 5, assuming that within every vector the curvature is constant. The time-dependent shape of the actuator is characterized by the matrix of angles. Each next angle along the columns is relative to the previous one, while the angles of the first column may be arbitrary and can perform well for representing the orientation of the clamps. Since the segments are independent, it is possible to interpolate the result along the coordinate obtaining a continuous curve of the actuator.

The image of the actuator is recorded by a camera and may be processed by some convenient image processing software or even manually with ruler and protractor. A proper lighting and exposure greatly simplifies the procedure of placing 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 consists of a single contrast curved line. Applying the procedure for sequential images gives a 3-dimensional evolution of curvature along the actuator in time. As an example, the graph given in figure 6 depicts the actuating-relaxing behavior of the actuator presented in figure 1.

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 discretization of the recorded image. Hence, it is important to provide sufficient resolution of the pixelated representation of the actuator to minimize image processing errors.

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| **Figure 5**. Vector representation of an actuator. | **Figure 6**. Transient behavior of an IPMC actuator. |

**4.2. Characterization of the bending moment of the bending actuator**

In cantilever mode – fixed end attached to contacts – the bending of the actuator is determined by the summarized moment of several components caused by the effects of osmotic, electrostatic and elastic stresses of the sample. When the sample is mounted edgewise, the component caused by the weight of the actuator in the particular environment in the plane of bending is assumed being zero. In the scope of the current paper, the mass and inertia of the actuator are neglected and we treat each time step as an independent static problem. According to the Euler-Bernoulli law the bending moment along a cantilever beam is proportional to the local curvature:

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where is the modulus of elasticity and is the second moment of area.

When the cross-section of samples is a rectangle, the second moment of area is expressed as

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where and are width and height of the cross-section of the sample respectively.

The described methodic does not pose any restrictions to the shape of the actuator, it works as well when the second moment of inertia is not constant along the sample e.g. the sample is tapered, or its cross-section differs from rectangular. In our case the summarized bending moment of the time-dependent behavior of the sample presented in figure 1 reflects exactly its bending depicted in figure 6, yet in a different vertical scale.

**5. Viscoelastic electromechanical model of IEAP actuators**

**5.1. Previous work**

The concept of the distributed electromechanical model of IEAP actuators is introduced in [DM]. This approach declares that the bending of an IPMC actuator at any time and any point is caused by the charge carried over between the electrodes at that point by that time. The amount of charge is determined by the inhomogeneous transitory voltage that in turn is uneven due the electrical resistance of the electrodes. The electrical constituent of that model depicted in figure 7 resembles a sophisticated lossy RC transmission line. The conductivity of the electrodes of the IEAP material is represented by a series of resistances of the opposite electrodes Ra and Rb, while the capacitance C, and the loss parameters G form the impedance of the material.



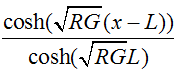
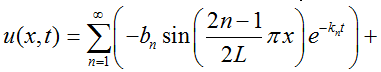
**Figure 7**. IEAP resembling a lossy RC transmission line. ***Siia Cauer!***

Although that paper emphasizes the flexure of the IPMC actuator is non-uniform in spatial and time domain and that it is scalable by means of the length of the actuator, it is contented with the simple proportional relation between the flexure and the charge, and discontinues just when the phenomenon of back-relaxation becomes noticeable. The objective of the current paper is to proceed from that stage of development.

One of the advantages of the approach of RC transmission line is the presence of the analytical solutions of its response to some specific input signals, particularly to the Heaviside step function. This enables easy comparison of the model with the real response of the actuator to the input voltage signal representing a flat step up. Moreover, it is possible to attach additional contacts to the surface of the actuator, and in this way validate the electrical parameters of the transmission line [XX, XX]. The propagation of voltage along this RC line is described by the following PDE:

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It is a pseudo-parabolic PDE, similar to those describing e.g. diffusion in porous media or heating a rod with radiation and loss present [XX]. Fortunately, it is possible to solve it analytically for utilizing the method of separation of the variables [DM, RCGWLine]. Moreover, by applying the initial and boundary conditions – initially the line is discharged and its other end is open – the analytical expression describing the time-evolution of the voltage distribution on the line after applying a voltage step to its input is

 (V2)

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where L is the length of the sample,

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and

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It turns out that the complicated time-evolution of the voltage distribution on the line is defined solely by the measurable parameters: by impedance of the material, by the length of the sample, and by time. Although the PDE describing the transient transferred charge is analogous to (10), it is impossible to apply the boundary conditions and obtain the solution, similar to (11). Instead, if initially the line is discharged, the charge is expressed via voltage as

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As an illustrative example, the simulated transient behavior of voltage and charge of the actuator depicted in Fig. 1., are given in Fig. 11.

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| (a) | (b) |
| Fig. 11. Voltage (a) and charge (b) profiles for a typical IPMC actuator. | |

**5.2. Distributed viscoelastic electromechanical model of IEAP actuators.**

The illustratory diagram of our viscoelastic electromechanical model of IEAP actuators is given in Fig. 12. It consists of infinite series of infinitesimally short lumped units, similar to those described in Section 3, while the conductive opposite electrodes of the IEAP material assure the electrical conductivity between the lumped units. Its electrical equivalent circuit is a lossy RC transmission line, described in the previous section, while the mechanical constituent stands of series of infinitesimally short hinged links. The buffers between the links are the models of viscoelasticity with internal strain, presented in Section 2. Even though the bending motion of IEAPs is caused by different physical processes, the correlation between the transferred charge and the input moments of the viscoelastic scheme is considered similarly linear by the coefficient as .



Fig. 12. The coupling between equivalent circuit and mechanical constituent.

Since we consider and I as constants, it is possible to group the coefficients can be grouped into . Now we can rewrite (3) for curvature as

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The curvature can be also transferred into Cartesian coordinates as

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where and are the initial position of the curve, and the angle

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where is initial angle at s=0.

**6. Verification of the model**

The first stage in the process of the verification of the model is the determination of the parameters. There are parameters of two types – electrical and mechanical. The electrical parameters include the resistance of the electrodes Ra and Rb, and the impedance of the material characterized by the capacitance C and two conductivities W and G. The mechanical parameters are the rate of relaxation λ=E/η, and the presented in section 5.2. While all electrical parameters are interpreted as per unit of length, a specific scaling parameter of this model is the length L of the sample.

The resistance of the electrodes can be measured with ohmmeter and 2- or 4-terminal sensing method, while the voltage should be kept within the electrochemical window of the electrolyte. It should be easy to characterize the impedance parameters C, W and G with a potentiostat. Unfortunately the parameters of the IEAP materials, especially the huge capacitance, exceed the range of the ordinary electrochemical equipment. As an alternative, these parameters were determined by fitting the step response of the propagation of voltage along the samples.

The whole set of the parameters was determined in 3 phases:

1. The resistances of electrodes Ra and Rb were measured using 4-terminal sensing method.
2. The actuator was excited with step voltage while the propagation of voltage along the actuator was measured by attaching 2-3 pairs of additional terminals onto the surface of the actuator, see [DM], The collected data was fitted against the equation (11) in order to obtain the values of C, G and W and assuming that and that Ra, Rb and L are known.
3. In Section 4 we demonstrated that for a short period after the step input voltage the difference between the behavior of strain and charge is inconsiderable. This presumption helps to determine λ and k by fitting the first few seconds of the experimentally recorded bending against the equation (15).

The fitting was performed using the differential evolution (DE) algorithm [X12]. The cost function was defined by simply minimizing the differences between measured and calculated voltages. Described two stage optimization gives an advantage to determine with coarse set of experimental points, while the C, G, W and are determined with fine set of points over shorter time period.

The validity of the viscoelastic electromechanical model of IEAP actuators presented in Section 5.2 was proved with three different IEAP materials:

1. The material **A** is a conventional wet IPMC with nafion membrane and platinum electrodes. It contains water; the cations introduced were Na+. Although it is intended for continous work in water environment, the measurements of short duration were passed in air. This material is soft, gains its maximal actuating amplitude in less than a second and relaxes back in about 10 seconds. Its working voltage should not exceed its electrochemical window – about 1.7V. The actuator presented in Fig. 1 is a typical example of this IEAP material.
2. The material **B** is a so-called carbon-polymer composite (CPC). Its membrane is made of a non-ionic polymer – PVDF, while the capacitance of the electrodes is contributed by the carbide-derived carbon. The whole laminate contains ionic liquid (EMIBF4) and is fabricated by hot-pressing. It is slow, gaining its ultimate bending amplitude in 60 seconds and relaxing back up to 600 seconds, but is much stronger than the other two. Its working voltage - up to 2.8V – is defined by the ionic liquid used. For details please refer to [doi:10.1016/j.carbon.2011.03.034].
3. The material **C** is an intermediate between the others. Its membrane is nafion and electrolyte is and ionic liquid EMITf, but the electrodes are made of carbide-derived carbon and covered with 15 um of gold. So, by means of membrane it is IPMC, but by means of electrodes it is CPC. The uppermost gold layer guarantees the good conductivity of the electrodes that in turn determines the speed and strength of this material. It gains its maximum amplitude in a few seconds and relaxes back in a few tens of seconds. Detailed description of fabrication and properties of **C** is available in [doi:10.1088/0964-1726/18/9/095028], referred to as the Carbon(1).

The parameters of the 3 materials are given in Table 1.

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|  | **A** | **B** | **C** |
| Dimensions | 45x3x… | 14x1x… | 28x5x… |
| R | 11,51 | 2509 | 2 |
| C | 0,00826 | 0,0088 | 0,035 |
| G | 0,001 | 0,000128 | 0,046 |
| W | 4,302 | 0,0075 | 0,0435 |
|  | 10 | 7,848 | 1,68 |
|  | 0,1776 | 0,00601 | 0,00269 |

**R, C, W, G, L, b, h, λ, k, μ, alfa .**

The parameter was derived from eq. 5 and was evaluated in region.

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| |  |  | | --- | --- | | Parameter | Value | |  |  | |  |  | |  |  | |  |  |   **Table 1. Measured parameters for CPC actuator.** | **Fig. 4 Measured and simulated voltage along the CPC at 2.0 V step input signal.** |

As seen in figure 1, good correlation between experimental data and simulated data with optimized parameters was obtained.

The simulated curvatures are in good correspondence with the actual curvature of the actuator. The maximum relative error over all simulations was less than 10%...

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| (a) | (b) | (c) |
| **Figure 2**. Simulated(above) and measured(below) curvatures of the samples **A**, **B** and **C** respectively. | | |



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| (a) | (b) |

Figure Y. Simulated and experimentally obtained shapes of bending of sample **A**.

**7. Discussion**

Miks me võime kasutada mudelit mitte-IPMC jaoks?

We have introduced a novel viscoelastic model for representation of bending motion of different types of IEAPs. The two equivalent forms of the model were derived out of which the first one was demonstrated as part of a distributed electromechanical model. However, there is no reason to underestimate the alternative form of the model which allows the substitution of the charge with corresponding current for instance.

Even though the model showed a good performance with the three actuators, there are still several issues to be concerned about:

* the distributed electrical model assumes unvarying resistance of the electrodes, however, it may not be justified for all IEAP actuators e.g. for PPy actuators;
* the deformation is modeled in accordance to Hooke’s law and is not covering the creep and the permanent deformation occurring beyond the elastic limit;
* using a single effective elastic modulus is appropriate only in the case of actuators with symmetric cross-section;

Interestingly, the super capacitors, CPC sensors, and CPC actuators – all have the same design. While for actuators, the bending of the laminate is a desired feature, it is highly unwanted in the case of supercaps. We believe that the same model might also contribute while describing the unwanted behavior of supercaps. There is also certain similarity between the behavior of our model and the CPC sensor output.

Sobib ka muuks. Mõtle näiteid. Näiteks on suur problem supercäppide liikumine. Indreku artiklis on öeldud et aktuaator, sensor ja supercäpp on ehituselt samad. Seega aitaks meie mudel kindlasti supercäppe modelleerida.

**Acknowledgments**

**Refs**

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X2: doi:10.1088/0964-1726/17/3/035011 Palladium buffer-layered high performance ionic polymer-metal composites

X3: doi: [10.1117/12.475167](http://link.aip.org/link/doi/10.1117/12.475167) Measurements and macro models of ionomeric polymer-metal composites (IPMC)

X4: doi: 10.1002/marc.201100535 Enhanced Biomimetic Performance of Ionic Polymer–Metal Composite Actuators Prepared with Nanostructured Block Ionomers

X5: doi: 10.1002/pen.21955 **New ionic polymer–metal composite actuators based on PVDF/PSSA/PVP polymer blend membrane**

X6: doi: [10.1063/1.2194127](http://link.aip.org/link/doi/10.1063/1.2194127) **Effect of solvents on the chemical and physical properties of ionic polymer-metal composites**

X7: doi:10.1088/0964-1726/20/8/083001 The state of understanding of ionic polymer metal composite architecture: a review

X8: doi: [10.1117/12.475201](http://link.aip.org/link/doi/10.1117/12.475201) Progress of experimental characterization and micromechanistic modeling of actuation of ionic polymer-metal composites

X9: PunnDistrModel

X10: doi: 10.1021/ma800956v

X11: Spie 2002 - 4695-33

X12: DOI: 10.1023/A:1008202821328

X13: doi:10.1088/0964-1726/18/11/115023

**Fraasid:**

as these parameters can be easily characterized by simple position sensors or force gauges

is of general-purpose and

Of course, the bigger number of vectors describes the shape more trustworthy, but technically it is As the

An already published technique, which satisfies the requirements described above , is to split the curve into segments or vectors (FIGX)[X13]. Although, this representation preserves the exact information about the endpoints of each segment, the curvature in between the points is reduced to a constant. Nevertheless, the desired accuracy can be reached by increasing the count of segments or decreasing the length of a segment.

Electrode resistance of the sample was measured experimentally … (directly/ four point method kõlaks siin hästi?) All other parameters were obtained by DE algorithm. Since the model is not yet fully optimized its evaluation in the meaning of reasonable precision is still computationally expensive. Therefore, the optimization of each sample was carried out in two stages. First, the parameters C, G, W andwere determined using fixed value of . In other words, we assume that at the beginning of the actuation there is no relaxation present. In second stage, all other parameters were fixed and the optimization was performed respect to only. Described two stage optimization gives an advantage to determine with coarse set of experimental points while the C, G, W and are determined with fine set of points over shorter time period.