IPMC Transduction Review

Abstract

This paper presents a review of using ionic polymer-metal composite (IPMC) materials as mechanoelectrical transducers. Recently more and more emphasis has been put on the research of IPMCs as displacement or velocity sensors for different applications. This has resulted in different theories and models to describe the mechanoelectrical transduction phenomenon. The paper gives an overview of the proposed transduction principles, developed models and the latest applications. In more detail, the history of IPMC materials, the physics and the electrochemistry behind the mechanoelectrical transduction, different black and grey box models, and novel real-world applications are discussed throughout the paper. However, despite of the latest advancements in the research of IPMC transduction, there is still some controversy in regard to some of the IPMC sensorial properties. For instance, it has been noticed by several authors that a signal delay which occurs in case of bending an IPMC. The general understanding of the physical principles about regular IPMC mechanoelectrical transduction is rather good. In the last section of the paper the novel results are presented for Cu-coated IPMC materials. Apparently the electrochemistry behind the transduction for Cu-coated IPMCs is significantly different. Besides the ionic diffusion, chemical reactions on the electrodes occur as well and dominate the actuation process. The preliminary experimental results show some promising opportunities for designing new Cu coated IPMC based sensors.

# Introduction

Ionic Polymer-Metal (IPMC) type of materials has been extensively researched during the last two decades. IPMC low voltage bending was first reported in 1992 by Oguro . From there on, many studies have considered IPMC electromechanical properties . The IPMC materials consist of a thin ionomeric membrane with the thickness of approximately 200 μm. The typical membrane materials are Nafion, Teflon, Flemion[7]. The membrane is coated with a thin layer of noble metal electrodes, such as platinum. To improve the electric conductivity of the electrodes, sometimes an additional layer of gold is added on the surface as well . As there are anions fixed to the polymer backbone, the membrane also consists of freely movable cations, so the overall charge of the material is balanced. Typical cations are Na+, Ka+, Li+ and Cs+ in water solution. The dry form of IPMCs has also been studied, where the ionic liquid (e.g TBA+) is used .

General conceptual design of IPMC was first described by Shahinpoor in 1992. The swimming robotic structure based on the IPMC actuators was proposed. The discussed kinematic equations were based on the work of Segalman et al, who presented a number of papers about modeling the IPMC materials . The diffusion equation describing the evolution of the solvent concentration and therefore the strain of polymeric gel material was proposed in 1991. In 1993 and 1994 Segalman et al also published the finite element analysis of the polymeric gel materials . In the following years attempts to formulate the electromechanical theory for IPMC materials were made. Shahinpoor presented a non-homogeneous large deformation theory of ionic polymer gels in electric and pH fields. The proposed model considered the spatial distribution of cations and anions inside the material due to the applied electric field. The model derived expressions relating the deformation as a function of electric field strength, dimensions and the material physical parameters [13-15]. In 2000 De Gennes presented the first phenomenological theory of sensing and actuation of IPMC. Nemat-Nasser and Li presented a modeling of electromechanical response of IPMC based on the electrostatic forces inside the IPMC. The cluster morphology of Nafion was also considered . In 2002, Nemat-Nasser stressed the role of hydrated cation transport within the clusters and polymeric networks of IPMC . Couple of years later, Weiland and Leo published a model, where the rotation of individual dipoles within a cluster was studied and related to the actuation of IPMC . In 2003 Nemat-Nasser presented an extensive study of actuation properties of IPMC with different membrane materials and type of cations . Typical Nafion based IPMC in most cation forms, when subjected to a small potential, undergoes a fast bending towards the anode, followed by a slow relaxation towards the cathode. However, When some large alkyl-ammonium cations are being used (e.g TBA+), the initial bending is rather gradual and the back relaxation is only partial[7]. Generally under DC, Nafion-based IPMCs do not maintain their initial displacement towards the anode and relax back towards the cathode. For some cations the back relaxation goes beyond the initial position [7, 20, 21]. SHOULD ADD FLEMION, ETC ???

In the recent years, research of the IPMC materials has been expanded more in terms of getting better efficiency, adaptability to environment, and applicability. Paquette and Kim investigated the low temperature behavior of IPMC and showed the material capability to operate even below -20C [22]. They also studied IPMC materials in a multilayer configuration and constructed an equivalent circuit model [23] . In 2006, Doyeon Kim and Kwang Kim presented an electrochemical analysis of IPMC. They showed that the performance degradation of IPMC over time is caused due to the Pt oxide formation and they also concluded from the analysis that the equivalent circuit of IPMC should include also inductor . In regard to IPMC applicability, Anton analyzed the usability of the material for real world applications and demonstrated the performance of IPMC using the inverted pendulum control . He also published a detailed description of the quasistatic mechanical behavior of IPMC actuator at large deflection and showed that a short actuator with rigid elongation behaves more linearly than a long one . In 2007, Kwang J. Kim et al. showed that IPMC operation in a saltwater environment is feasible and possible naval applications could be considered . At the same year, Dogruer et. al showed that when modeling an IPMC subjected to an aqueous environment, the hydrodynamic forces do not significantly affect the performance of the IPMC. In the following years, Doyeon Kim and Kwang J. Kim conducted an extensive study of IPMC materials exhibiting self oscillations . The mechanism behind this phenomenon was electrochemical reaction on Pt electrodes of an IPMC which was immersed in H2SO4 solution. A year later, Pugal et al published a finite element model to describe this phenomenon . Recently, Dogruer et. al showed that IPMC could be use for energy harvesting purposes. A study how to increase the efficiency is still in progress . A comprehensive research in regard to characterization of IPMCs for power harvesting was reported by Brufau-Penella et al. in 2008. The contribution of that research was a generic model that works with the IPMC material in dehydrated conditions.

##  IPMC materials as mechanoelectrical transducers

As there are quite many papers about IPMC actuation, the number of papers considering IPMC mechanoelectrical, or sensory properties is much smaller. However, during the last decades, several authors have focused their research on IPMC sensory properties and papers which consider IPMCs as sensor have been published[37-53, 53-58] . The first paper about ionic polymer sensors was published by Sadeghipour et al. in 1992. The system where pressure was applied to the thickness direction of platinum coated Nafion was described. The Nafion was not hydrated but was previously saturated with hydrogen. As the pressure was applied the system generated measurable voltage output. The study brings out that Nafion-base smart materials could successfully be used as vibration sensors due to the high sensitivity and linear characteristics. Couple of years later, Shahinpoor reported new discovery of a new effect in ionic polymeric gels - the ionic flexoionic polymeric gelectric effect. It means that IPMC strips created the output voltage when was subjected to flexing or loading. Later Shahinpoor and Mojarrad investigated sensing of tip displacement of IPMC strip in cantilevered configuration. They observed that the output voltage was dependent on the orientation of the transducer with respect to the electrodes. In 1998 the first review paper of IPMC materials as biomimetic sensors and actuators was published by Shahinpoor[62]. As was brought out above, De Gennes presented the phenomenological theory of sensing and actuation of IPMC in 2000.

The principle of sensing is reverse of the actuation. When the material is bent, some of the solvent carrying charged ions are mechanically forced to the vicinity of one electrode. So there forms excess of charges on the expanding side of the material. This in turn results in electric field across the polymer thickness and the corresponding voltage signal can be detected on the electrodes [20, 40, 45, 48, 63]. Bonomo et al. have done some characterization of IPMC sensors . Their research suggests that output voltage is roughly linear in deformation and nonlinear behavior seems to occur only in particular working conditions - this is rather case for lower frequency mechanical input. One of the advantages of IPMC based sensors over their actuation properties is that the wet environment is not the key issue for IPMC-based sensors to work well. In fact they do not suffer the same drawbacks as actuators do . Chen et al. reported also weak sensing after IPMC was taken out of water due to the excess water . However their research indicates that the amplitude of the sensing signal increased to the maximum at about t=5 minutes, and it started to decrease afterwards. The time-varying response is believed to arise from water evaporation of the IPMC sample. Water evaporation also increases Young modulus, but it is not the dominant factor of sensing response change. So the idea that hydration level changes sensing behavior through the influence of ionic diffusivity still needs some research.

Several configurations have been proposed to create IPMC based sensor-actuator systems. Nakadoi [52]has proposed an integrated IPMC actuator-sensor system on a single film and examined the electric interference from actuator phase to sensor phase. Chen et al. have proposed a system where an IPMC sheet is coupled with two polyvinylidene fluoride (PVDF) film[64], bonded to the sides of the IPMC. The authors have overcome the feedthrough problems that they had with the given configuration in past . The paper brings out that the differential configuration adopted in both sensors was critical in eliminating feedthrough coupling, rejecting sensing noises induced by thermal drift and EMI, compensating asymmetric tension/compression responses, and maintaining structural stability of the composite beams. For the first time feedback control of IPMC was successfully demonstrated using only integrated sensors showing that one can simultaneously regulating/tracking the bending displacement and monitoring the force output (or vice versa). The actuator/sensor system was verified by measuring forces during penetration of soap bubble. However, only small deformation of IPMCs was assumed. For the large displacements the discrepancies between the bending displacement obtained from the PVDF and from the laser sensors were evident. Ideas of using the same IPMC strip as a sensor and as an actuator have been also published. For instance, Punning [53]has demonstrated a self-sensing actuator. The sensing was based on the voltage drop measurements - half of the IPMC was fixed and another half was able to move and there was an extra wiring at the tip of the IPMC to measure voltage drops. The fixed part provided reference voltage and the voltage of the deforming part was subject to the voltage change due to surface resistance variations. The measured voltages provided some information about bending direction and extent of the sheet. For the given model to work, a surface resistance study was also carried out . As the surface resistance acted to the bending as a delicate feedback, more the resistance changed, more the sensory output of the given system got disturbed. Also Farinholt , Konyo[43], and Bonomo have noted that more the sensor strip is crooked, the more the output signal gets disturbed. Specifically, there exists a certain time delay between the deformation and sensor output. The authors have not discussed the reason for this phenomenon, so more research would be required to investigate the behavior. However, Punning has proposed a circuit model which predicts the delay.

Despite the fact that there is not as much research done about IPMC sensory properties as about the actuation properties, there are already some applications proposed for IPMC sensors . What makes IPMC sensors appealing is their linear output voltage. Bonomo has suggested that the output voltage is not strong enough to produce nonlinear phenomenon. [Bonomo06] At the same time the current output is shown to scale linearly with the width of the IPMC and inversely with the length, which allows developing simple IPMC based sensors. Bonomo [57]has introduced a prototype of a probe for biomedical applications. The proposed device consists of an IPMC actuator and an IPMC sensor and it is able to recognize the presence of tissues coming into contact with its sensitive part. Also a model which corresponded well to the experimental data was developed. Biddiss has analyzed IPMC materials for using as sensors in hand prostheses. The results look promising and the authors brought out that contrary to many traditional approaches, IPMCs do not require external power supplies or auxiliary mechanism, making them suitable for this kind of applications. However some parameters such as temperature variation and its influence on the sensitivity were not measured above 50C. Chen has demonstrated an application of IPMC/PVDF sensor - micro-injection of living Drosophila embryos. IPMC/PVDF structure and sensing circuit was used. Experimental results showed that the developed IPMC/PVDF sensor-actuator system, together with a model-based compensation algorithm, can perform effective, simultaneous actuation and sensing. Paola has proposed a vibration sensor of an IPMC in cantilever configuration. The single degree of freedom system was modeled with spring/dampener equivalent model. The results showed that IPMC could be used as a vibration sensing transducer with adequate sensitivity and linear characteristics.

When it comes to modeling of IPMC actuation, there are mainly two types of models available - physical models which consider material properties add baumgart, etc and circuit equivalent models , which are based on the electrical measurements. Both types of models for IPMC mechanoelectrical transduction properties have also been developed. Bonomo brings out the fact that the voltage generated by the sensing is not sufficient to produce nonlinear effects, therefore linear approximation in models could be used. Noteworthy is the fact that the hydration level was discovered to play a significant role for sensors and the sensing behavior improves when the hydration level is in equilibrium with the environment. In the paper also a circuit based model for an IPMC sensor was proposed. On the other hand, a dynamic, physics-based model is presented by Chen for ionic polymer–metal composite (IPMC) sensors. The model was an infinite-dimensional transfer function relating the short-circuit sensing current to the applied deformation. It was obtained by deriving the exact solution to the governing partial differential equation (PDE) [44, 70]for the sensing dynamics, where the effect of distributed surface resistance was incorporated. The model involves basic electrostatics and ion migration/diffusion description and goes beyond that. First, it incorporates the effect of the distributed surface resistance, which is known to influence the actuation and sensing dynamics . Chen's model comparison with results shows that on the magnitude plot, both with surface resistance and without, the results are same. On the other hand, the phase plot with surface resistance is more accurate. An exact, analytical solution to the PDE was obtained by converting the original time-domain equation to the Laplace-domain version. Also instead of limiting to step deformation only, an arbitrary mechanical deformation stimulus was allowed, which is of interest for real applications. The result of the work was a physical model, which also takes into account the surface resistance. However there also were some discrepancies between the model description and the experimental measurements. The author proposed that those could have been because of non-modeled nonlinearity.

Another highly physical model was proposed by Farinholt . The author makes similar hypothesis as was made by Nemat-Nasser and Li that the charge density at the surface of the polymer is proportional to the induced stress. The assumption was utilized by them to model the actuation properties of the material. The basic premise of the model is predicted by a field formulation that includes nonzero ion flux. This assumption permits a solution to the equations that represents a short circuit condition in which the measured current is related to the time rate of change of the charge density at the surface of the polymer.

# Mechanoelectrical transduction mechanisms of IPMC

There are several different ways how the physics behind the mechanoelectrical transduction of IPMC is understood. Tadokoro, et. al{{108 Tadokoro, S. 2000}} proposed a model of ionic polymers based on ionic motion. The electromechanical transduction is explained with the forces imposed on the ions as they migrate in the polymer due to an external electric field. Electrostatic interactions produced by ionic motion were also modeled. De Gennes et al, Asaka and Oguro {{44 de Gennes,P.G. 15; 81 Asaka,Kinji 2000; }} explained the underlying causes of the transduction with solvent fluxes and pressure gradients. Under the electric field the counter ions drift, and they carry with them a certain number of water molecules. When the molecules pile near the cathode, a local overpressure is created, this in turn causes deformation in the membrane. There are two forms of transportation inside the polymer: charge transport (with a current density ) and solvent transport (with a flux ). The standard Onsager relations for such system are {{107 De Groot, S.R. 1966}}

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Here, is the membrane conductance, is the Darcy permeability and is a cross coefficient. The induced electric field is proportional to an applied bending torque , which is the cause of the pressure gradient in the material. Now, when a mechanical torque is applied to the material, there will be a finite water flux , but electric current remains zero. The magnitude of induced electric field can be calculated using

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where is Poisson’s ratio and is the thickness of the membrane. This model, though, ignores the electrode effects and also the osmotic pressure contribution.



Figure induced electric dipole within a cluster. The dipole is proportional to cluster radius , distance between the ions and inversely proportional to imposed radius of curvature of IPMC [Nemat Nasser and Li 2000]

The different approach to describe the mechanoelectrical transduction is to consider the electrostatic interaction within the polymer. The model was proposed by Nemat-Nasser and Li [Nasser and Li 2000, Nemat-Nasser 2002]. The underlying cause of actuation is explained by the internal stresses induced by interaction between ion pairs inside a cluster. The main components of the stress are the electrostatic stress due to the charge imbalance in the polymer and the osmotic stress, which is due to redistribution of water molecules. The governing equation for counter ion distribution due to applied electric field is:

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where is counter ion concentration, D is diffusion constant, is electric field potential, is fluid pressure, and free water velocity. The term is a combination of water and counter ion molar volumes. The above equation together with an applied time independent electric field gives the stress field inside the polymer as follows

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where is anion density, is a constitutive parameter, which relates the charge density to the electrostatic stress, andis the osmotic stress. Using the same ionic cluster material approach, the induced electric field due to imposed curvature is calculated as well. The electric field is caused by induced electric dipoles in the clusters and the overall field strength is proportional to the sum of dipole moments. The approximate expression without explicit constants is as follows:

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where is electric dipole and is the distance between the clusters. The electric dipole of a single cluster is illustrated in Figure 1.

When Nemat-Nasser and Li charge sensing model is about ionic dipoles induced in the polymer clusters, Farinholt and Leo, by using the same field equations, presented a hypothesis that the charge density on the surface of the polymer is proportional to the induced stress. This is basically the reverse of actuation model of Nemat-Nasser and Li. The model, though, ignores the convective term of Eq. (3) By assuming linear relation between the stress and the charge density, the induced electric field inside the polymer due to a tip deflection is proportional to the terms and :

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Where and are inverse time constants. The induced electric field at one instant of time after the mechanical deformation is illustrated in Figure 2. The electric field profile is almost identical to the case when external voltage is applied on IPMC.

The aforementioned models are usable for mechanoelectrical transduction problems. Even though the approaches are different, the practical value for different models remains and lots of effort has been put in to explain the physics behind the phenomenon. However, there are some areas of mechanoelectrical transduction of IPMC, which remain unclear.

Figure Normalized electric field, calculated using the model proposed in [Farinholt]

Different authors have obtained more phenomenological characteristics of IPMC sensors. [Bonomo06, characterizati, PUNNING07-1, konyo2004dvs, Farinholt04] For many cases, there exists a certain time delay between the imposed displacement and the measured voltage of the IPMC. The experimental data suggest that the output signal depends on imposed curvature of IPMC as illustrated in Figure XXX. [PUNNING07-1]. Also somewhat contradicting characteristic of IPMC mechanoelectrical transducer is the stability of the measured output signal. Some experiments suggest that the IPMC characteristics do not change after hours of work and measured voltage is still reliable. [Bonomo06]. Although the wet IPMC produces noisy signal at first, but after the material reaches the state of equilibrium, the signal remains noiseless during many hours of work. [sdarticle] For other instances, the sensor signal started to show decrease of the amplitude after only of work [Chen07]. Those areas, though well observed, need some more study to better understand and possibly improve the material.

## IPMC mechanoelectrical transduction nonphysical models

The physical models of IPMC transduction mechanisms are very advanced and provide insight to the physical processes behind the phenomenon. However, for practical use, those models might be too complicated. Therefore simpler grey-box models have been developed to control and register the outputs of IPMC transducers.

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