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Investigation on the ion diffusion and migration transport mechanism in Polypyrrole tri-layers actuators conducting polymer

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Ion transference in conductive polymers has been initiated to rely equally upon material characterization and investigational circumstances. In the field of actuators, conductive polymers have been utilized with great success. This study employs the electrostatics module of the COMSOL Multiphysics finite element model to enhance existing modeling techniques and to comprehend the ion diffusion and migration transport mechanism in layered conductive polymers, specifically Polypyrrole tri-layers actuators. The model controls the specific flow pattern and temporal advancement of electrical voltage, cationic and anionic concentrations. The model uses only material characteristics to predict structural deformation in tri-layer actuators, indicating its potential practical utility. Moreover, it is sample-independent, making it valuable for electro-active polymer applications. Evaluation using existing data demonstrates that the simulation's predictions align with high certainty across the entire range of assessed input potentials, further validating its reliability and effectiveness.

Keywords: ion diffusion; ion migration; conducting polymer; Polypyrrole tri-layers actuators; tri-layer actuators; COMSOL model.

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Introduction

Metals are excellent conductors of electricity and widely accessible. However, manufacturing them into intricate shapes is challenging and often expensive. In contrast, thermoplastics offer the opposite advantage. They can be easily and inexpensively molded or extruded into various shapes and sizes at a low cost. Additionally, the specific weight of typical industrial plastics is approximately one-third that of comparable metal materials. Hence, electrically conductive plastics are frequently preferred for various applications where metal conductivity is not essential. Conductive polymers and plastics prove suitable for a wide range of diverse applications [1]. Previously, the issue of accumulating electrostatic charges was addressed by incorporating carbon-black or acetylene-black into regular rubber and metal powders and extending this method to plastics. Another approach involved amalgamating metal threads or metal-coated fibers with carbon-based fibers to overcome this propensity. [2]

Polymeric compounds capable of conducting both electricity and heat were developed through the incorporating of larger amounts of conductive ingredients into plastics and elastomers [3]. Conductive polymers garner significant attention within the realm of polymer science due to their unique structure and chemical properties. They exhibit exceptional conductivity [4], ranging from S cm^{-1} to 500 S cm^{-1}, which can be significantly enhanced up to 3065 S cm^{-1} through copolymerization or pretreatment procedures. [5] The band-gap energy in conducting polymers is relatively small, Usually falling within the range of 1 to 3 electron volts (eV), in contrast to the nearly 10 eV gap found in traditional polymers [6]. Conducting polymers exhibit various mechanical properties, such as mechanical elasticity, which is a significant advantage for designing technical devices [7] using different processing methods. These polymers are derived from inexpensive sources like aniline, pyrrole, thiophene, and their byproducts, through simple chemical or electrochemical polymerization processes. Conductive polymers prove to be exceptional options for electronic applications, including electromagnetic interference shielding and flexible "plastic" transistors. Additionally, they find applications in electro-optical devices, such as electro-luminescent polymer spectacles, light-emitting diodes (LEDs), and solar cells. Additionally, they find applications in electrochemical devices like rechargeable batteries, corrosion inhibitors, and chemical and biochemical sensors [8-10]. MacDiarmid and Heeger, have previously verified that thin pieces of "doped" polyacetylene can change sun light to electricity and therefore possess possible applications especially as a solar converter along with for batteries [11]. Doping has notably amplified the potential effectiveness of conductive polymers by increasing their electrical conductivity by several magnitudes. This involves the introduction of electron acceptors (p-type) such as iodine or arsenic pentafluoride, leading to substantial improvements in conductivity [12].

The electrical conductivity exhibited by polyacetylenes, polyphenylene, and polyphenylene sulfide can be significantly influenced by the doping technique [13], involving the addition n-type material as electron doners such as sodium or sodium naphthalene. This process has a notable impact on the electrical conductivity of these polymers. During the mid-1970s, conductive polymers surfaced as an innovative category of organic materials exhibiting electrical properties similar to those of metals or inorganic semiconductors. At the same time, they retain the typical features of traditional polymers, such as simplicity in synthesis, processing simplicity, and pliability in handling [14]. Conducting polymers can be produced through chemical polymerization and/or electro polymerization, each method presenting its own set of advantages and drawbacks. Chemical polymerization can be scaled up to yield large amounts of a product, yet typically requires catalysts that are not easily separated. The purification of the polymer is a time-consuming process. Alternatively, electro-polymerization offers some useful features, for instance lack of catalysts, the direct formation of the polymer film onto the surface of the electrode and ease of controlling film width [15]. Furthermore, electro-polymerization allows for characterizing the plated film in situ using electrochemical methods. However, it is not easily scalable for mass production purposes [16].

Temperature plays a crucial role in the ion transport mechanisms of Polypyrrole (PPy) tri-layer actuators, influencing both diffusion and migration in several ways. Ion diffusion in PPy follows the Arrhenius relationship, meaning that as temperature increases, the diffusion coefficient rises exponentially due to increased thermal energy [17]. This leads to faster ionic motion, reducing response time and improving actuation speed. Also, ionic conductivity in the electrolyte layer improves with temperature due to lower viscosity and enhanced ion mobility. This strengthens the migration contribution to ion transport, especially at higher applied voltages, where electric field effects dominate over diffusion [18]. Temperature fluctuations can lead to thermal expansion and contraction of the polymer matrix, affecting ion pathways and polymer-electrolyte interactions. At lower temperatures, rigidity increases, potentially restricting ion mobility and slowing actuation response. Elevated temperatures can accelerate side reactions, such as unwanted redox processes, polymer oxidation, or electrolyte decomposition, reducing device lifespan. Optimizing operational temperature ranges is essential to balance high ionic mobility with chemical stability for improved actuator performance [19].

Many researchers argue that ion transportation in polymers is not solely determined by the electric field, as polarons/bipolarons form stable electrostatic interfaces with ions when they exist within the polymer [20]. Osteryoung and Pickup, however, suggested that diffusion alone cannot adequately account for experimental findings, particularly under the influence of high potentials [21, 22]. Apart from relocation, ion transmission in conducting-polymers is not implicit yet. Fickian diffusion, wherein ion transport occurs due to concentration gradients, has been widely presumed in models that depict ion migration within conductive polymers [22], disregarding the fact that dispersion in polymers might be influenced by the polymer moderation procedure [22], and disregarding the potential impact of the polymer moderation process on dispersion [23, 24]. In this study, the main emphasis is on introducing and developing a novel integrated Multiphysics finite model for the Polypyrrole trilayer actuation device, eliminating the need for sample-specific constraints.

I. Theory

In nonappearance of fluid turbulence, ions are transported from solution to an electrode by three major mechanisms; migration, diffusion, and convection [25]. Migration exerts a significant influence on ion transference. Migration refers to the transfer of charged particles within an electric field, determines the velocity of charged particles according to the equation $v = \mu \times E$, where, μ represents mobility (cm/sec. V), and E represents the electric field (V/m). This approach is employed by utilizing the electrostatic interface of COMSOL Multiphysics, incorporating both diffusion and migration transfer mechanisms while ignoring convection as outlined in formulas 1 and 2. The model computes charge transfer as defined by the Nernst-Planck relation, considering the response time, *t* through migration and diffusion processes:

$$\frac{\partial C_k}{\partial t} = \nabla \cdot (D_k \nabla C_k + z_k \mu_k F C_k \nabla \emptyset) \qquad (1)$$

Where, *k* point and *F* represents the Faraday's constant. "k" is represented by "A" in the setting of distribution hole in the C-P province, "k" is represented as "H", the movable cationic types in the solution, "k" is represented by "C”, "k" is denoted as "Ck". Additionally, types of the coefficient of diffusion are represented as "Dk", the types' valence charge quantity is denoted as "zk" and ionic mobility is represented by "μk". The symbol "φ" represents the resident electrical potential. The electrical potential with the resident remaining charge-density (ρ) is related by Poisson's relation, as stated by:

$$\varepsilon_o \nabla . (\varepsilon_r \nabla \emptyset) = -\rho = -F \sum_k z_k C_k \qquad (2)$$

where ε_o and ε_r represents the free space permittivity and the average dielectric constant, respectively. Though the passing natural surroundings of the design is mainly suitable for expecting actuator reaction time. A precise exemplary of charge transference also improves some necessity for connected current observing characteristic [8].

The exemplary design for charge transference comprises three sub-domains, with each one representing a separate sheet of the actuator separately. *CP* layer width is h_p = 50 μm and the spongy film essential width is h_m = 110 μm in agreement with available investigational confirmation information. The electrical potential, cation attention, anion attentiveness, and hole attentiveness are all controlled by the model, along with their precise allocation and gradual advancement. The 1-D The model operates under the assumption that the profiles of potential and concentration remain unaffected by the actuator distance. This hypothesis is predicated on the claim that these effects are lessened near the layer C-P boundary where there is a conductive underlayer.

It can be stated that selecting appropriate boundary conditions is crucial for ensuring reproducibility and accuracy of the results. Typically, the boundary conditions for ions and charges in different layers are chosen based on the physical, electrochemical, and mechanical behavior of the actuator. The boundary conditions ensure that the total charge in the system remains conserved. At the interfaces between the electroactive layers and the solid electrolyte layer, continuity conditions are imposed for electrical potential to maintain electrochemical equilibrium. Also, the ion flux at the boundaries is controlled by electrochemical kinetics and diffusion constraints. Typically, no-flux boundary conditions are applied at the edges to prevent artificial ion loss. By implementing these well-defined boundary conditions, the study ensures consistent migration behavior across different trials, leading to high reproducibility of the experimental and simulated results.

Interface circumstances are specified for the electrostatic boundary, comprising an electrical crush and a functional voltage (electric potential) on the borders, along with external conductors as depicted in Fig. 1.

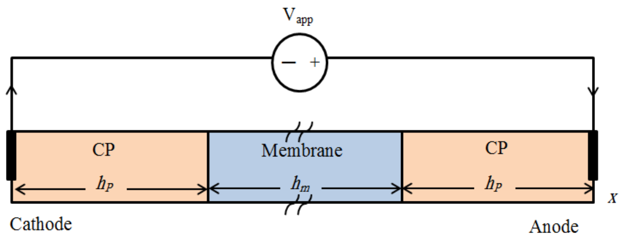

Fig. 1. The boundary circumstances performed in the anticipated charge transference exemplary.

As shown in Figure 1, flux boundary scenarios are also used to simulate the passage of holes between CP layers via the external circuit. These configurations encompass the dimensionless structure of Ohm's law, which connects current density and the inherent electrical field intensity, while also assuming the existence of locations with negative charges for the migration of holes. This facilitates the achievement of charge neutrality within the specified regions. The remarkable characteristics of experimentally detected anions are showcased in the exemplification through the necessity of an occupation on a flattened stage.

The intended output seamlessly transitions from one to zero when employing the overload portion methods. The alteration region has a thickness of $D_{mol}/\ (C_{A0,electrolyte} + D_{mol})$, in which D_{mol} signifies the molar attentiveness of allowed anionic charge transporters assumed through $D_{mol} = D/F$, and D is the net charge. The permissible highest concentration was specified as $C_{A,electrolyte,max}$ = 6 mol · l^{-1}. The function controls the incoming flow by multiplying the species' mobility with the function's output. This effectively restricts concentration increases that arise from migration once the maximum redox charge is attained. The same method was applied in the CP domain to decrease the ionic mobility of both anionic and cationic species as the solubility limit is approached. This helps alleviate physically extreme concentration gradients.

II. Results and discussion

The study helped resolve several long-standing debates in literature. The first key question was whether migration contributes to ion transport in conjugated polymers. This is a crucial issue, as it directly pertains to the fundamental driving mechanisms in these materials and significantly impacts their controllability in devices. Results in this study provide clear evidence that migration must be considered, as none of the diffusion-only models tested could adequately explain the experimental observations. Notably, the transport mechanism shifts from being diffusion-dominated at low overpotentials to migration-dominated at higher voltages, where ion movement is primarily governed by the locations of voltage drops within the system.

The second question concerned the presence of net charge in the polymer. Interestingly, the simulations indicate that this factor is largely irrelevant-whether or not a net charge exists has no significant impact on the overall transport behavior. Figures 2 to 5 explain the temporary solution to the transport issue under an applied potential of 0.8V. The simulation results are qualitatively compared with experimental data for an ion-barrier-covered film during electrochemical reduction. The model effectively captures the key behaviors observed, including the formation of a frontal side of the potential profile. Results showed qualitative behavior reminiscent of what Madden proposed [26], it also recalls migration impacts within the charge transport simulation as suggested elsewhere [27]. By simulating the geometry described in [26] and [27], the authors of this investigation are able to qualitatively compare the results of the simulations with the experimental results. It can be stated that, the study findings are inconsistent with the experimental results [26]

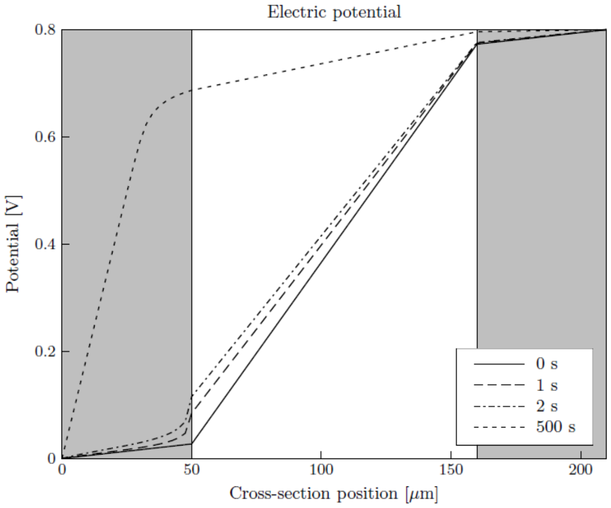

Fig. 2. Simulation result demonstrating the transitory progress of the electric potential profile in reaction at 0.8 V.

Figure 2 illustrates the transient progress of electric potential. The initial profile reproduces the expected potential drops consistent with the specified layers dielectric constant. The adverse potential motivates cations and holes toward the cathode, on the other hand, anions are directed toward the anode. Reductions in electrical charge and the development of growth layers occur at the junction between the C-P layers and the core of membrane, where there are variations in the rates of transportation within the neighboring fields. Due to the decreased mobility of the cations compared to their anionic counterparts, there's an anomaly observed in their concentration patterns. This anomaly is similarly manifested in the electric potential profile.

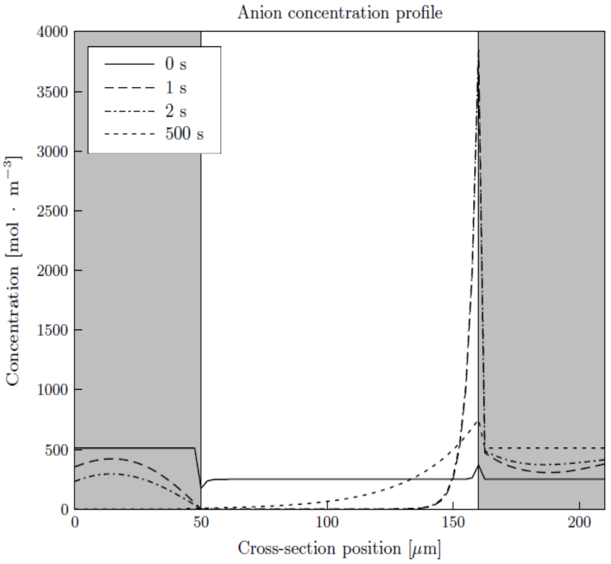

Fig. 3. Transitory progress of the negatively charged ions concentration profile in response to 0.8 V applied voltage.

The concentration profile of the anionics illustrated in Figure 3 is particularly noteworthy. Meanwhile, the comparatively mobile anionic species primarily contributes to the generated mechanical stress. Whitin the cathodic layer, the starting concentration includes anions released through redox exchange induced by the applied voltage, along with the preliminary concentration of negative ion in the electrolyte. Over time, the applied voltage pushes the negative ion towards the anode, progressively decreasing the presence of the negative charges at the boundary between the cathode and the membrane. The recombination of holes at the cathode, combined with the limited mobility of cations, further decreases anions at the interface between the cathode C-P layer and the electrode, ensuring the maintenance of local charge neutrality.

The hole transfer illustrated in Figure 4 specifies two specific behaviors in the C-P layers. In the cathode C-P layer, a non-zero preliminary hole density rises from the statement of prompt release of holes when an electric potential is applied. Meanwhile, the concentration of holes gradually decreases at the border of the membrane of cathode. As holes recombine with electrons from the external circuit, their concentration diminishes, leading to a corresponding generation of equivalent holes in the anode membrane. The recently created holes travel towards the cathode under the influence of the applied electric field as well.

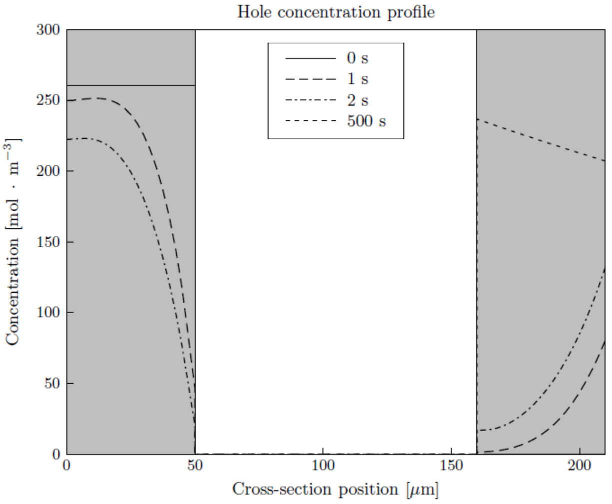

Fig. 4. Transitory progress of the concentration of hole response to a potential of 0.8 V.

The spreading of cation species is presented in Figure 5. Due to the relatively stationary nature of cations, notable concentration gradients arise at the interfaces of layers when subjected to an electric field. This results in a faster reorganization of both positive and negative ions. The delayed reaction of the holes strengthens the detected behavior where the movement of anions predominantly influences the actuation response that negative ion motion controls the response of actuation, particularly with the rising operational frequency observed in continuous flapping scenarios. Meanwhile the strain induced by local charge density makes the model viable to anticipate actuator's minimal latency.

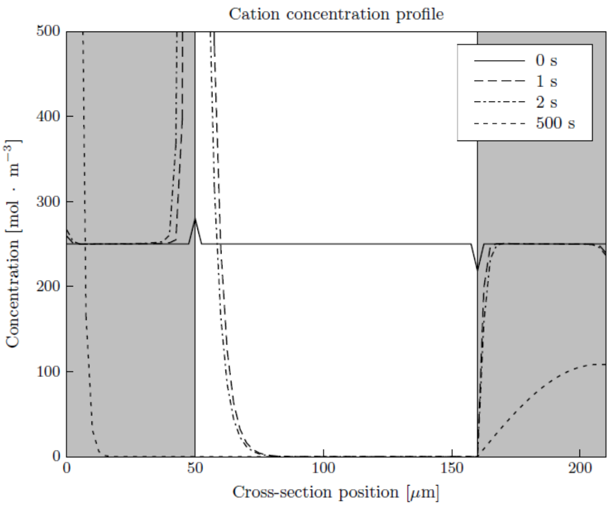

Fig. 5. Transitory progress of the positive ions' concentration with response to a potential of 0.8 V.

Conclusions

The electrostatics module of COMSOL gained awareness utilizing the COMSOL multiphysics software. The objective of the model is to generate actuator deflection curves corresponding to various applied potential. Results imply that, mechanical curvature consistently fall within 96% confidence intervals across the entire range of input possibilities assessed when compared with reported data. The predictions of mechanical curvature align with highly dependable intervals across the entire spectrum of evaluated input voltages. This study described the existence of CP actuators modeling methods and recognized that their usage for predictive design is incomplete. This is more likely attributed to their reliance on properties specific to the specimen, which must be experimentally taken into account or observed in its actual environment. The model predicts the physical distortion of the tri-layer-actuators based on the properties of the material, aiming for sample-independent applicability. The proposed model demonstrates that the voltage and ions concentration profiles are not influenced by the actuator length. Moreover, the model expects that the structural irregularity for tri-layers actuators over material properties might possess suitable application such as an electro-active polymer design capability.

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Дослідження механізму йонної дифузії та міграційного транспорту в тришарових поліпірольних актуаторах з провідним полімером

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Ініціація транспорту йонів у струмопровідних полімерах дозволяє у рівній мірі покладатися як на характеристики матеріалу, так і на методи дослідження. У галузі актуаторів все більше використовують провідні полімери. У цьому дослідженні застосовано модуль електростатики скінченно-елементної моделі COMSOL Multiphysics для вдосконалення існуючих методів моделювання і розуміння механізму дифузії та міграції йонів у шаруватих струмопровідних полімерах, зокрема, в тришарових поліпіролових актуаторах. Модель контролює специфічну картину потоку і часовий прогрес електричної напруги, концентрацій катіонів і аніонів. Модель використовує лише характеристики матеріалу для прогнозування структурних деформацій у тришарових актуаторах, що вказує на її потенційну практичну користь. Крім того, вона не залежить від зразка, що робить її цінною для застосування в електроактивних полімерах. Оцінка з використанням наявних даних демонструє, що прогнози симуляції збігаються з високою точністю в усьому діапазоні оцінених вхідних потенціалів, що додатково підтверджує її надійність та ефективність.

Ключові слова: дифузія йонів; міграція йонів; провідний полімер; Поліпірольні тришарові актуатори; Модель COMSOL.