PAPER

Biomimetic polypyrrole based all three-in-one triple layer sensing actuators exchanging cations†

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Simultaneous actuation and sensing properties of a triple layer actuator interchanging cations are presented for the first time. Thick polypyrrole (pPy)/dodecylbenzenesulfonate (DBS) films (36 µm) were electrogenerated on stainless steel electrodes. Sensing characteristics of pPy-DBS/tape/pPy-DBS triple layer artificial muscle were studied as a function of electrolyte concentration, temperature and driving current using lithium perchlorate ($LiClO_4$) aqueous solution as electrolyte. The chronopotentiometric responses were studied by applying consecutive square waves of currents to produce angular movements of $\pm 45°$ by the free end of the triple layer. The evolution of the muscle potential (anode film *versus* cathode film) during current flow is a function of the studied chemical and physical variables. The electrical energy consumed to describe a constant angle is a linear function of the working temperature or of the driving electrical current, and a double logarithmic function of the electrolyte concentration. Those are the sensing functions. The cation exchanging bending triple layer actuator senses the working conditions. Similar sensing functions were described in the literature for devices interchanging anions. Irrespective of the reaction mechanism, a single electrochemo–mechanical device comprised of two reactive polymer electrodes (oxidation film and reduction film) works simultaneously as both sensor and actuator (self-sensing actuators). These are the general sensing properties of dense and biomimetic reactive gels of conducting polymers. Thus, any reactive device based on the same type of materials and reactions (batteries, smart windows, actuators, electron–ion transducers) is expected to sense surrounding conditions, as biological organs do.

Introduction

Biological functions are based on chemical reactions taking place in living cells. For the last few years man-made technology is trying to produce materials mimicking biological composition, reactions and functions. Among those materials reactive gels composed of conducting polymers (CPs), water and ions are the simplest biomimicking materials.[1–6]

Interactions of polymeric chains with the electric fields or electric currents provide new electromechanical and electro-chemomechanical properties and devices. Those devices can produce mechanical energy from electrical energy[1–11] or from electrochemical energy,[12–14] using polymer films. In analogy with the natural muscular functions (electric pulse, ionic exchange, chemical reactions, conformational movements and generation of mechanical energy), devices based on those interactions are named as artificial muscles.

The flow of an electric current through films of CPs in the presence of electrolytes (solid or liquid) promotes electrochemical reactions leading to volume changes.[15–19] From the viewpoint of electrochemical kinetics, any physical or chemical variable acting on the reaction rate should produce a change in the electrode potential when the reaction occurs under constant current in accordance with the rate equation. Under those conditions the potential evolution of the reactive material is expected to be a sensor of the surrounding chemical or physical conditions. Thus, conducting polymers can be regarded as soft, wet, actuating and sensing materials.[20] They mimic most of the properties of biological materials.

Polypyrrole films exchanging anions during reactions have demonstrated to sense temperature and concentration variations in the ambient environment.[21–27] Bilayer and triple-layer artificial muscles constructed from those conducting polymer films can sense any change in the same variables and/or the weight of any trailed material.[28–31] Similarly, tactile muscles capable of sensing

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† Electronic supplementary information (ESI) available: Fig. S1, scheme for experimental bilayer sensing actuation; Fig. S2, chronopotentiogram of the triple layer; Fig. S3–S5, variation of muscle potential evolution while describing different angles under studied variables. See DOI: 10.1039/c1jm13374h

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the presence of an obstacle on its way have been developed. These devices can provide information about the mechanical resistance of an obstacle to be pushed and shifted.[32,33] During working, the actuating signal (i.e. the current) is sent to the device through the two connecting wires and the sensing signal (the muscle potential) is followed simultaneously between the same connecting wires. Thus, devices based on chemical reactions are simultaneous actuators and sensors capable of providing a new technological paradigm.

For those CP artificial muscles having a prevalent exchange of cations during reactions, volume changes and movements produced by the same current are in opposite direction to those produced in CP artificial muscles exchanging anions.[31,34,35] The material swells during reduction by entry of cations and shrinks during oxidation by expulsion of cations, whereas conducting polymers exchanging anions swell during oxidation and shrink during reduction. Recently we have demonstrated that both the pPy-DBS films and the bilayer artificial muscles pPy-DBS tape can sense, while reacting, surrounding physical and chemical variables.[36,37] There, the device potential was followed using a reference electrode. Moreover, the system requires three electrodes: an actuator working electrode (WE), a metal counter electrode (CE) and the reference electrode (RE) (ESI, Fig. S1†). This configuration produces practical difficulties in developing devices, wasting most of the consumed energy. The energy consumed by this muscle is a small fraction of the overall electrical energy as most of the consumed energy is required to drive the electrolyte discharge at the metal counter electrode.

Here we check and characterize, for the first time, simultaneous actuating and sensing properties of an all three-in-one actuating device, a triple-layer CP/tape/CP device, in which the CP films exchange cations with the electrolyte during actuation. CP films on either side of the tape can act as working electrode and counter electrode, respectively, and the reference electrode is short-circuited to the CE (Fig. 1). The muscle potential (potential of the working CP film versus the potential of the counter electrode CP film) can be followed during current flow, i.e., during working. Another advantage is that the integrated electrical energy will include the full energy consumed by the muscle: that is required to oxidize one of the CP films plus the energy required to reduce the second film.

Experimental

Chemicals

Pyrrole (Fluka®) was purified by distillation under vacuum using a diaphragm vacuum pump (MZ 2C, SCHOTT®) and stored under nitrogen atmosphere at −10 °C. Anhydrous lithium perchlorate salt (Fluka®) and dodecylbenzenesulfonic acid (DBSA) solution (70 wt% in 2-propanol, Aldrich) were used as received. Ultrapure water from Millipore Milli-Q equipment was used.

Film preparation

pPy films were prepared at room temperature (22 ± 1 °C) in dark conditions in a one-compartment electrochemical cell using 50 mL of an aqueous solution containing 0.2 M DBSA and 0.2 M pyrrole monomer. The working electrode was an AISI 316 stainless steel plate, with a thickness of 1.24 ± 0.01 mm and having a surface area of 2 × 5 cm ± 0.05 cm on each side. Deposition was performed on both sides of the immersed part of the electrode having an area of 2 × 3.3 cm ± 0.05 cm on each side. Two large electrodes having an area of 2.5 × 5.5 cm ± 0.05 cm of the same material were used as counter-electrodes. They were symmetrically placed at a distance of 1 ± 0.1 cm from the working electrode to obtain a uniform electric field. A standard Ag/AgCl (3 M KCl) electrode from Metrohm® was used as reference electrode.

The pPy film was electro-generated by applying a constant anodic current density of 0.75 mA cm^{-2} for 100 min. The overall charge consumed during the electro-polymerization was 60 C and the polymer weight was 54.5 ± 0.01 mg. Once generated, the pPy film was polarized on the stainless steel working electrode at 0.5 V for 4 min in order to obtain an intermediate oxidation level. The polypyrrole film from the steel electrode borders was scraped. Two separate films were obtained (coating each of the electrode faces), with a mass of 26.42 ± 0.1 mg each, determined by means of a precision balance (Sartorius®, SC2 Model) of ±0.1 μg precision. After peeling off from the working electrode, the films were immersed in de-ionized water for 48 hours in dark conditions to remove excess DBSA from the polymer surface and de-ionized water was changed every 7 hours, otherwise the film loses the adherence to the tape and the devices stop working. Finally, pPy-DBS films were air-dried at room temperature. Films of thickness 36 ± 1 μm (measured using a COMECTA® electronic digital micrometer) were used for experimentation.

Fabrication of the triple layer

The pPy-DBS/tape/pPy-DBS triple layer actuator was fabricated using a commercially available double sided non-conducting polymeric tape from 3M and by sticking each of the two pPy-DBS films on either side of the tape. The two pPy-DBS films have
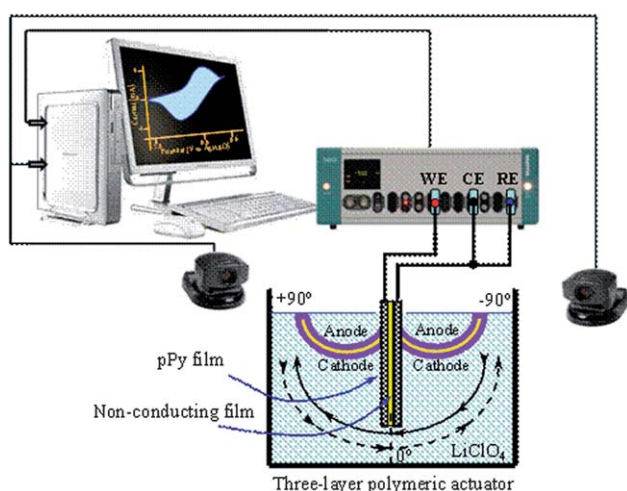

Fig. 1 Scheme of the electrochemical cell and configuration of electrodes (WE, working electrode; CE, counter electrode; RE, reference electrode) used to study the electrochemical actuation and sensing behaviour of the triple-layer actuator (pPy-DBS/non-conducting tape/pPy-DBS). A vision system was used for video recording and to measure the angular movement in aqueous electrolyte solution.

an intermediate state of oxidation and have dimensions of 2×0.5 cm ± 0.05 cm, thickness of 36 ± 1 µm, and weighing 3.3 ± 0.01 mg each. One side (pPy-DBS film) of the triple-layer actuator works as anode and the other side acts as cathode. No electrical contact exists between the two conducting polymer films due to the non-conducting tape present in-between. One of the pPy layers was connected to the working electrode output of the potentiostat–galvanostat. The pPy film attached to the opposite side of the triple layer was connected to the counter electrode output of the potentiostat–galvanostat. This side of the triple layer was short circuited with the reference electrode. The free end of the triple layer actuator described a continuous angular movement around the fixed end. The whole experimental setup was immersed in aqueous electrolyte solutions and characterized by applying square current waves, and recording the chronopotentiometric responses.

Characterization

All electrochemical studies were performed using an Autolab PGSTAT-100 potentiostat/galvanostat controlled by a personal computer using (GPES®: General Purpose Electrochemical System) electrochemical software. Temperatures were controlled by using a Julabo T25 cryostat/thermostat having a precision of 0.1 °C. All the other experiments were performed at room temperature.

All the experiments related to the sensing characteristics were performed in a one compartment electrochemical cell. In all the experiments, the triple-layer actuator was held by a non-conducting alligator clip with metal contacts adhered separately on both sides of the clip. They allow independent contacts with the working electrode film and with the counter electrode film of the muscle. The electrochemical measurements were carried out in aqueous solutions of LiClO$_4$. The muscle potential evolution for sensing characterization was recorded while the muscle described a constant angle of $\pm 45°$. The angular rates were calculated from images taken by a vision system using EVI-D31 SONY® digital cameras controlled by a Matrox® card and a control system programmed in C++ for image processing in Matlab®. The experimental setup is shown in Fig. 1.

Results

Voltammetric response

The electroactive material in the pPy-DBS/tape/pPy-DBS triple layer actuator was characterised by cyclic voltammetry, short circuiting both pPy films and using a stainless steel plate as the counter electrode and an Ag/AgCl (3 M KCl) as the reference electrode. The cyclic voltammogram (CV) was recorded between the potential limits of -0.8 V and 0.3 V at a scan rate of 5 mV s^{-1} in 0.1 M LiClO$_4$ aqueous solution at room temperature. The resulting CV showed the presence of one anodic maximum at 0.047 V and a cathodic maximum at -0.547 V (Fig. 2). Before starting sensing experiments, the triple layer was stabilized by subjecting the two polypyrrole films, simultaneously, to 10 consecutive potential cycles between the specified potential limits. The potential sweep was stopped at -0.1 V during the anodic voltammogram in order to get an intermediate oxidation
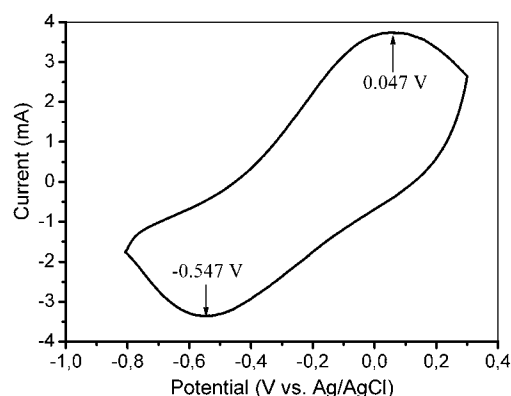

Fig. 2 Voltammogram recorded for pPy-DBS layers (short circuited) from the triple-layer actuator at a scan rate of 5 mV s^{-1} between the potential limits of -0.8 V and 0.3 V at room temperature (electrolyte: 0.1 M LiClO$_4$ aqueous solution).

state for the subsequent actuation under constant current: oxidation of one film and reduction of the second film.

Actuation and sensing characteristics of the pPy-DBS/tape/pPy-DBS triple layer actuator

Actuation and sensing characteristics of the pPy-DBS/tape/pPy-DBS triple layer actuator were studied under galvanostatic conditions (Fig. 3). Before studying those characteristics, the triple-layer was allowed to undergo an initial polarization by applying a constant electric current of -0.01 mA for a period of 250 seconds. It was then submitted to consecutive square waves of electric current by applying $\pm i$ mA for the required time in order to describe a constant angular movement of the free end of the triple layer. The input signals and the resulting output chronopotentiograms when the pPy-DBS/tape/pPy-DBS triple
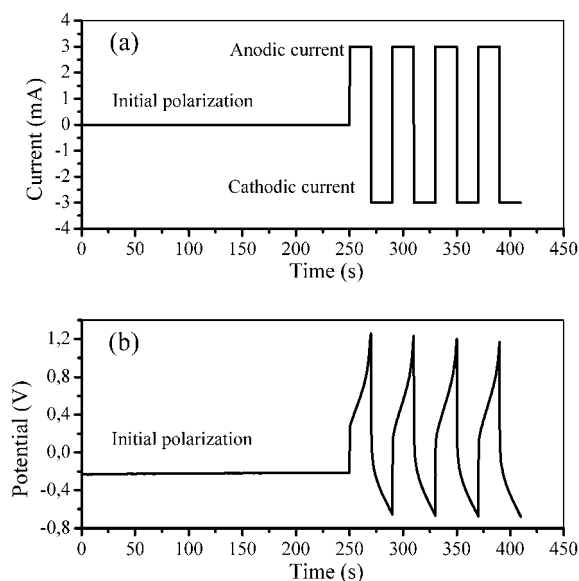

Fig. 3 (a) Square current waves applied to the working electrode of the triple layer and (b) resulting chronopotentiometric responses showing the evolution of the muscle potential (working electrode *vs.* counter electrode).

layer actuator was subjected to ± 3 mA of current for a period of 20 seconds (consuming 60 mC: $Q = It$) in 0.1 M LiClO$_4$ aqueous solution at 25 °C are given in Fig. 3(a) and (b), respectively. Sensing characteristics were monitored as a function of: applied current, temperature and different concentrations of the electrolyte. The third chronopotentiometric cycle was used for the sensing analysis of the studied variables. Here we consider that the currents are positive or negative, respectively, for the anodic or cathodic currents that flow through the working electrode. Under those conditions the free end of the triple layer describes an angular movement of $\pm 45°$, starting from the vertical position to move back and forth (Fig. 4).

pPy-DBS/tape/pPy-DBS triple layer actuator senses current

For studying the dependence of the applied current on the chronopotentiometric responses, the experimental procedure described in Fig. 3 was repeated by applying different currents ranging from ± 0.75 mA to ± 15 mA for different time periods so as to keep a desired continuous angular movement of $\pm 45°$ by flow of a constant charge of ± 60 mC. The experiments were carried out at a room temperature of 25 °C in 0.1 M LiClO$_4$ solution and the chronopotentiograms were recorded (ESI, Fig. S2†).

Fig. 5 shows the stabilized third anodic and cathodic chronopotentiograms. Higher currents induce higher potential steps at the starting point, due to different types of resistance associated with the triple layer such as: film reaction resistance, charge transfer resistance, solution resistance or counterion diffusion resistance. After this initial step, the potential gradually increases. This evolution takes place at higher potentials for increasing anodic currents, following the electrochemical reaction of polypyrrole (Fig. 5(a)). It can be seen that the potential
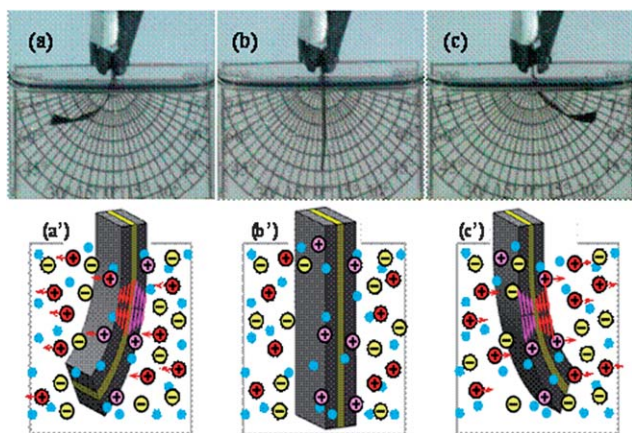
Fig. 4 Bending movements (amplitude, ± 60 degrees) of the triple layer pPy-DBS/tape/pPy-DBS in 0.1 M LiClO$_4$ aqueous electrolyte. During anticlockwise movement, from (a) to (c), the left side pPy-DBS film swells by reduction (flow of cathodic current) and the right side film shrinks by oxidation under anodic current flow: (c') shows exchange of cations and the generated stress gradients. By reversing the direction of the current flow, the left side film becomes the anode and the right side the cathode and the clockwise movement is generated from (c) to (a): ionic exchange and stress gradients are reversed (a').
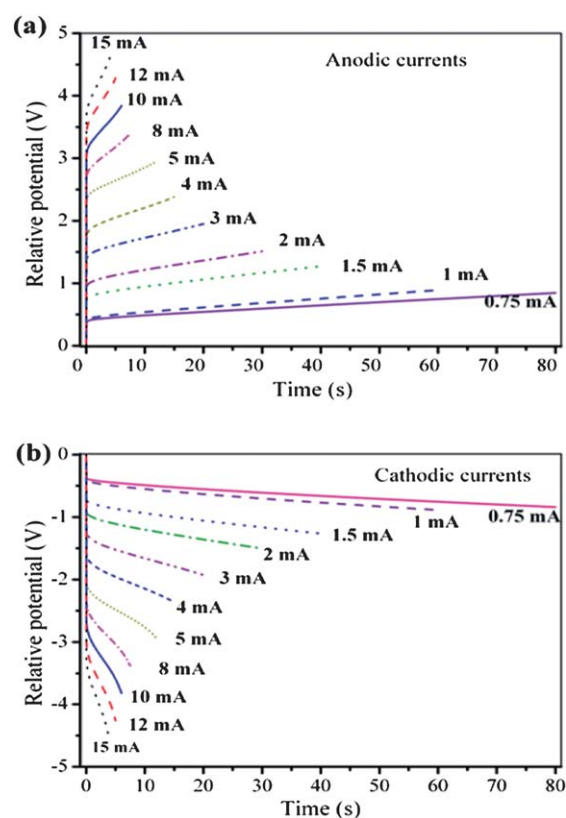
Fig. 5 Current sensing characteristics of the triple layer actuator. (a) Anodic and (b) cathodic chronopotentiograms for an angular movement of $\pm 45°$ under different applied anodic and cathodic current square waves (electrolyte: 0.1 M LiClO$_4$ aqueous solution).

gradually decreases towards higher negative potentials for the increase of cathodic currents (Fig. 5(b)).

The muscle potential (potential difference between anodic and cathodic pPy films in the device) of the triple layer actuator for describing angular movements of $\pm 15°$, $\pm 30°$, and $\pm 45°$, respectively, was obtained from the chronopotentiograms. The muscle potential follows three linear relationships (each one by considered angle) with the applied current. Same kind of potential evolutions were observed for the anodic (ESI, Fig. S3a†) and cathodic (ESI, Fig. S3b†) processes. Slopes (mV mA^{-1}) are the increase of muscle potential when the magnitude of the driving current increases by one unit. Slopes are found to increase for increasing constant angles: with the increase of time period of the applied current reactions (oxidation or reduction) become more resistive for higher oxidized or higher reduced states of the conducting polymer.

pPy-DBS/tape/pPy-DBS triple layer actuator senses temperature

By following the experimental procedure described in Fig. 3, the temperature dependence of the chronopotentiometric responses, from 5 °C to 45 °C, under flow of square potential waves of current ± 3 mA in 0.1 M LiClO$_4$ describing a constant angular movement by the free end of the triple layer of $\pm 45°$ was studied (Fig. 6). Fig. 6(a) and (b) show the attained anodic and cathodic responses, respectively. Anodic and cathodic currents flow for
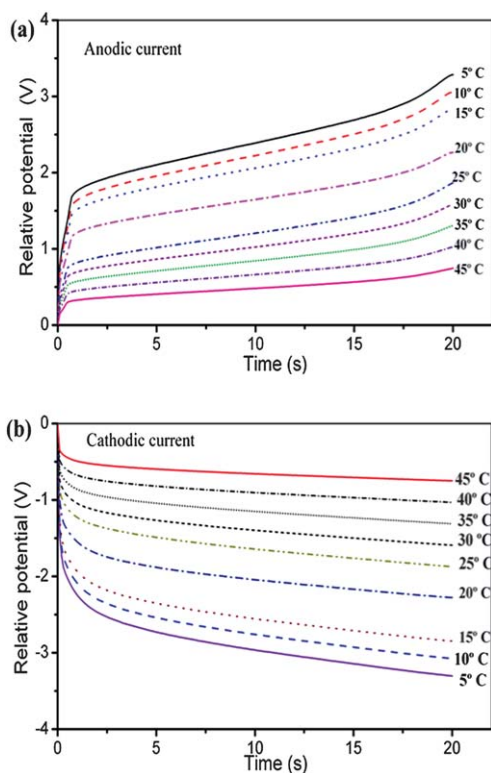
Fig. 6 Temperature sensing characteristics of the triple layer actuator. (a) Anodic and (b) cathodic chronopotentiograms obtained during a constant angular movement of $\pm 45°$ of the triple layer actuator under a constant current flow of ± 3 mA at different temperatures (electrolyte: 0.1 M LiClO$_4$ aqueous solution).

the same time of 20 seconds for all the experiments and consume ± 60 mC for both the forward and backward movements.

Considering Fig. 6(a) and (b), the potentials at three different times of $t_1 = 6.66$ s, $t_2 = 13.33$ s and $t_3 = 20$ s correspond to angular movements of $\pm 15°$, $\pm 30°$ and $\pm 45°$, respectively. These potentials have a linear relationship with temperature (ESI, Fig. S4a and b†). Whatever be the considered angle, the muscle potential decreases for higher experimental temperature, as expected from the Arrhenius expression for the reaction coefficient. Slopes represent the decrease of the muscle potential when the temperature increments are one degree centigrade. Increasing slopes are obtained for increasing described angles: reactions (oxidation or reduction) become more resistive for more advanced reaction states.

pPy-DBS/tape/pPy-DBS triple layer actuator senses electrolyte concentration

The chronoamperometric responses were studied for different electrolyte concentrations of aqueous LiClO$_4$ solutions, ranging from 0.001 M to1 M, at 22 °C, by subjecting the three-layer to a constant current of ± 3 mA, and allowing it to describe constant amplitude of the angular movement of $\pm 45°$. It can be inferred from reaction (1) (see Discussion) that rising electrolyte concentrations facilitate the reaction to occur: under constant current lower potentials are expected during the movement as observed from the results shown in Fig. 7(a) and (b).

The muscle potential and the electrolyte concentration have a semilogarithmic dependence (as expected from the Buttler–Volmer expression) for the same oxidation states attained at $t_1 = 6.66$ s, $t_2 = 13.33$ s and $t_3 = 20$ s, which correspond to angular movements of $\pm 15°$, $\pm 30°$ and $\pm 45°$ respectively (ESI, Fig. S4a and b†).

Discussion

The general shape and the peak potentials in the CV of the short circuited films of the triple layer are similar to the one recorded for pPy/DBS films in previous results.[19,31] A large potential separation of about 0.6 V between the peaks is a consequence of the slow diffusion of balancing counterions through extremely thick pPy films. By integration of the anodic and cathodic branches of the voltammogram we obtain the involved charge in the films oxidation, Q_{ox} (218.5 mC) and the involved charge in the films reduction, Q_{red} (216.9 mC). Those charges and the pPy weight, w, give the specific charge, Q_{ms} (33.106 C g^{-1}). This is equivalent to a maximum variation of the specific concentration of exchanging cations in the film, $[C^+] = Q/(F \cdot w)$, of 3.4312×10^{-4} mol g^{-1}, where F is the Faraday constant (e$^-$ mol^{-1}). If during actuation of the device those magnitudes overcome their maximum value, the device moves outside its reversible redox behaviour and will progressively degrade (over oxidize).

The direction of angular movement of the pPy-DBS/tape/pPy-DBS triple layer actuator indicates that the polypyrrole film
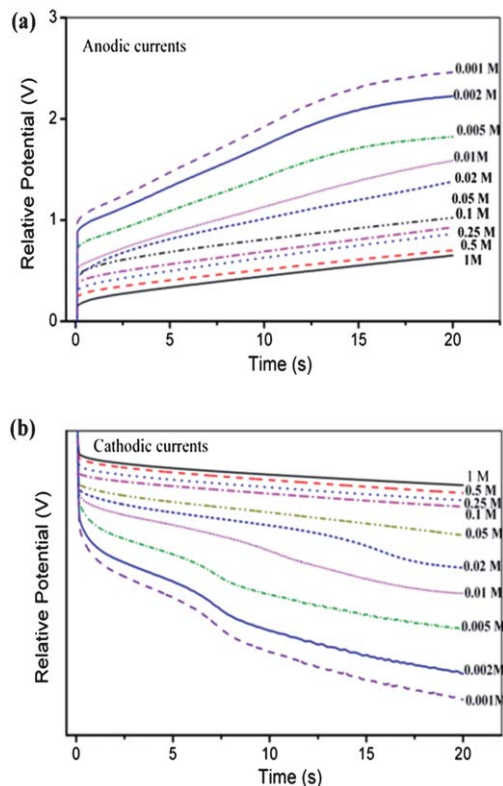

Fig. 7 Concentration sensing characteristics of the triple layer actuator. (a) Anodic and (b) cathodic chronopotentiograms obtained during a constant angular movement of $\pm 45°$ of the triple layer actuator under a constant current flow of ± 3 mA at various electrolyte concentrations (electrolyte: LiClO$_4$ aqueous solution).

acting as the cathode pushes the device and that acting as the anode trails the device as indicated in Fig. 4. It means that the pPy-DBS film acting as the cathode swells during reduction pointing to the insertion of cations as the prevalent ionic exchange between the film and the solution. The pPy-DBS film acting as the anode shrinks during the oxidation driven by the flow of the anodic current and due to the prevalent expulsion of cations into the solution.[19,34] We can conclude that the electrical current flow drives the redox (reduction–oxidation) processes exchanging the Li^+ ions between the polymer matrix and the electrolyte. This ionic exchange together with the solvent molecules is required for osmotic pressure balance. The bulky counterions, DBS^- ions, get trapped inside the polymer matrix.[19] The driving electrochemical reaction, excluding the water exchange, can be summarized as:

$$[(pPy)_{solid}(DBS^-Li^+)_n] \leftrightharpoons [(pPy^{n+})(DBS^-)_n]_{solid}$$
$$+ n(Li^+)_{solvent} + n(e^-)_{metal} \qquad (1)$$
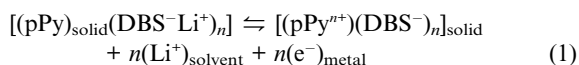
The reaction occurring from left to right is the anodic process (oxidation) in which electrons are extracted from the polymeric chains and the reverse process is the reduction.[1,6] The macroscopic change of volume induced by the continuous variation of the film composition promotes increasing stress gradients (of compaction and expansion at the anode and cathode, respectively) across the conducting pPy-DBS/non-conductive tape/pPy-DBS interfaces. As a consequence, the free end of the triple-layer actuator describes a continuous angular movement (Fig. 4).

The triple layer structure with one of the films acting as the anode and the second as the cathode under a constant applied charge per unit of time corresponds to a bending battery involving the charge/discharge of $Q_s = 9.09$ C g^{-1} to carry out a continuous angular displacement of $\pm 60°$ or $Q_s = 13.636$ C g^{-1} for $\pm 90°$. In the future, a good control system will allow recovering this charge and a fraction of the involved energy. Our working conditions are very stable because we move far away from the limiting conditions for the working films of Q_{ms} (33.106 C g^{-1}), that means far from the degradation conditions.

The exchange of cations by pPy/DBS materials, bilayers and triple layers is known from the literature. In the present study with the triple layer actuator, we have now quantified the involved charges, defined the limiting specific charges or concentrations to avoid degradation and have quantified the charge that can be recovered (the triple layer is a battery) during backward movement. In addition, the driving electrochemical reaction mentioned above (eqn. (1)) includes simultaneous sensing possibilities that had not been studied yet, for cation exchanging triple layers. Any physical or chemical variable acting on the reaction rates (oxidation or reduction) must induce changes in the muscle potential when driven by currents. Potential evolution and the current flowing during a given time are the magnitudes that define the consumed electrical energy in electrical motors ($E_e = QE$). It means that the muscle while working is expected to detect any variable (concentration, temperature, pressure, mechanical stress, presence of obstacles) by changing the muscle potential and by adapting to the required electrical energy.

The electrical energy consumed to describe angular movements of $\pm 45°$ by flow of the studied anodic or cathodic currents is $E_e = I \int E dt$, where I is the applied current, E is the potential, and t is the time. The integral, $\int E dt$, represents the surface area under the experimental chronopotentiograms. Fig. 8 shows that the electrical energy consumed for describing an angular displacement ($\pm 45°$ here) is a linear function of applied current. The slopes 3.51 J g^{-1} mA^{-1} and -3.39 J g^{-1} mA^{-1} represent the increment of consumed specific energy when the applied current increases (for anodic and cathodic processes respectively) by one mA, quantifying the ability of this particular actuating material to sense the driving current. Those results indicate that the triple layer actuator can sense, while working, any change of the driving current. One device with only two connecting wires (Fig. 1) accommodates both actuating (current) and sensing (potential) signals. The device is a sensing-actuator or sensing muscle.

By integration of Fig. 6(a) and (b), the electrical energy consumed by the muscle working at different temperatures is obtained. Fig. 9 shows a linear decrease of the electrical energy consumed to describe a constant (45°) angular displacement, when the temperature increases. The slopes -0.52 J g^{-1} $°C^{-1}$ and -0.42 J g^{-1} $°C^{-1}$ of the anodic and cathodic, respectively, currents represent the increment of the consumed specific energy when the temperature decreases by one degree centigrade. Those slopes quantify the sensing ability of this actuator material for temperature variations. Results depicted in Fig. 9 suggest the ability of our triple layer device to sense temperature while working.

By integration of the experimental curves from Fig. 7(a) and (b), Fig. 10 is obtained, showing that the electrical energy consumed to describe a constant angle, under flow of a constant current for a constant time, increases for decreasing electrolyte concentrations. Thus the triple layer actuator is proved to work as a concentration sensor while working. The oxidation and reduction slopes -0.23 (J g^{-1} M^{-1}) and -0.23 (J g^{-1} M^{-1}) indicate the increment on the consumed energy per unit concentration variation, quantifying the sensing ability of this actuator material for electrolyte concentration variations.

The electrochemical reaction (1) is the common source of both actuating and sensing properties. It produces conformational
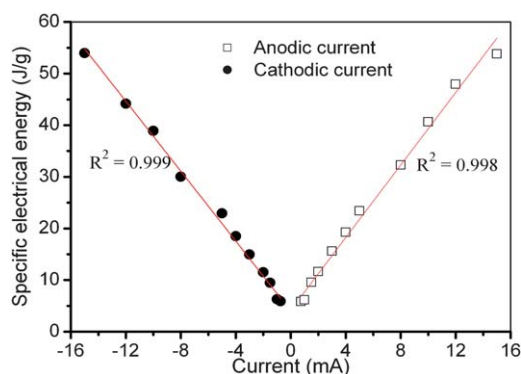

Fig. 8 Linear variation of the consumed electrical energy with different applied currents for (a) anodic and (b) cathodic currents corresponding to the experimental results shown in Fig. 5(a) and (b) during current sensing characteristics of the triple layer actuator.
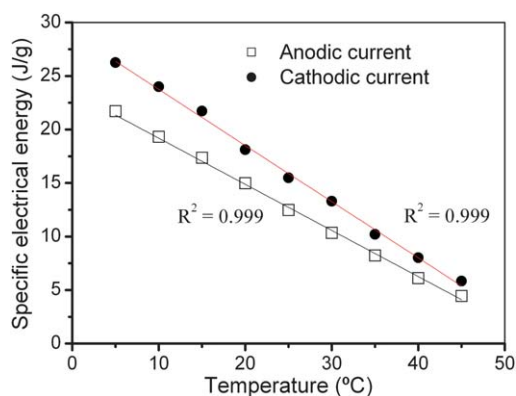
Fig. 9 Linear fit of specific electrical energy consumed with different temperatures during the anodic and cathodic processes corresponding to the experimental results shown in Fig. 6(a) and (b). The polypyrrole weight in the actuator was 6.6 mg. R^2 is the correlation coefficient of the linear fit.
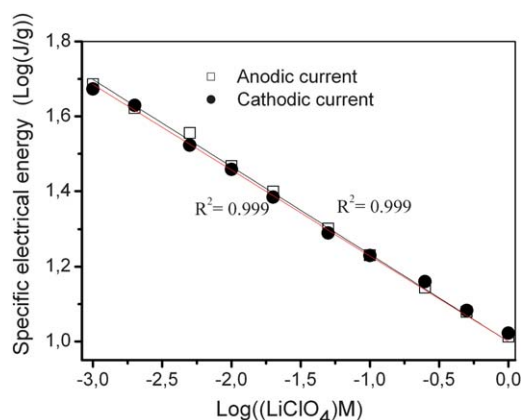

Fig. 10 Variation of specific electrical energies (obtained by integration of the current responses corresponding to Fig. 7(a) and (b)) with the logarithm of the experimental concentration of the electrolyte. The weight of the polymer film was 6.6 mg. R^2 is the correlation coefficient of the linear fit.

movements of the chains, with generation or elimination of free volume and macroscopic volume changes. As for any electrochemical reaction the driving current is sensed by the resistances, including the reaction resistance, opposed to the current flow. Since the electroactive component in the device is the reactive pPy films, we suggest that the sensing ability of the muscle potential is offered by polypyrrole reaction.[18,20–24,28,31–35] As reported earlier for free standing films, bilayer or trilayer actuators, or for any hybrid films based on conducting polymers exchanging anions with the solution, the ability of the muscle potential to sense the change of any physical or chemical variable acting on the reaction rate is a general property of the reactive soft and wet gel materials, exchanging anions or exchanging cations during the driving reaction. Therefore, we suggest that this is a general principle that can be applied to any electrochemical device (actuator, battery, smart membrane, and so on) based on the Faradaic reactions of conducting polymers or any other reactive gel.

Those shifts of the consumed specific energy as a function of the experimental variables must influence the efficiency of energy conversion from chemical energy into mechanical work. This aspect will be studied in a subsequent work using artificial muscles shifting different masses.

Conclusions

The bending movement and the subsequent angular displacement of the free end of the pPy-DBS/tape/pPy-DBS triple layer device are controlled by the electrochemical reaction (1) promoting a prevalent exchange of cations with the electrolyte. Results presented in this work unambiguously corroborate that, while working, the triple layer electro-chemo-mechanical actuator, similarly to anion exchange triple layer actuators, can sense working and surrounding conditions such as driving current, electrolyte concentration or working temperature. Magnitudes as consumed electrical energy or muscle potential allow the obtention of the calibration equations for each sensing property. The consumed energy gives calibration equations with higher correlation coefficients than 0.99. Every slope (consumed energy per unit of the sensed magnitude and per unit of pPy/DBS mass) is a characteristic of the conducting polymer constituting the triple layer.

Only two connecting wires (Fig. 1) include both actuating (current) and sensing (potential) signals. The device is a sensing-actuator or sensing muscle mimicking natural muscles. In the dark we can extend our arm to catch an object and our brain knows, immediately, the exact energy required for its displacement.

The fact that both sensing and actuating properties are based on the same electrochemical reaction allows stating a general principle: any device based on the electrochemical reaction of the pPy/DBS material (artificial muscle, battery, smart membrane, smart window, smart drug delivery, electron/ion transduction, and so on) can sense both, working and surrounding variables, while working.

Acknowledgements

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