Electronic Memory Effects in a Sexithiophene−Poly(ethylene oxide) Block Copolymer Doped with NaCl. Combined Diode and Resistive Switching Behavior

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Electrical transport in devices consisting of an electrode of a conducting polymer (poly(ethylenedioxythiophene):poly(styrenesulfonate), PEDOT:PSS); a layer of a sexithiophene−poly(ethylene oxide) (6T-PEO) block copolymer with an Al top electrode is investigated. These devices show diode behavior with the larger current density flowing when the PEDOT:PSS electrode is biased positive with respect to the Al electrode (forward bias). Introduction of inorganic salt (NaCl) in the PEDOT layer results in resistive switching behavior under forward bias while retaining the diode character. The switching allows for storage of information and rewritable memory operation is demonstrated for the diodes although the retention time of the information is still very short (∼10 s). The reported combination of switching and diode behavior is an important requirement for passive matrix addressing of resistive switching memory cells in an array and shows that materials with combined ion and charge transport properties are interesting for information storage.

Introduction

The possibility of using π-conjugated polymers in electronic memory elements is currently being explored by a large number of research groups.[1] One of the approaches being followed is to make diode-like structures for which the resistance at a particular “read” voltage can be modified by application of a higher voltage.[2] One of the attractive features of these diodes is that they may be integrated into a matrix array of plastic memory elements relying on passive matrix addressing to read out the individual cells.[3] Forrest and co-workers have used a polymeric fuse in combination with an inorganic diode to make a passive matrix array of write-once read-many memory elements.[4] The incorporation of a diode is required for selective writing and reading of individual cells in the array, suppressing cross-talk between the cells in the matrix array. Recently, they showed that also an inorganic−organic heterojunction can serve as a diode in memory cells.[2b] To come to *rewritable* memory cells that can be integrated into a passive matrix array, many requirements have to be fulfilled (see also Supporting Information). Important features of such memory elements are to exhibit diode character and have two different resistance levels in forward bias.

It has been shown that the electrical properties of π-conjugated polymers doped with mobile ions can be modified by application of bias voltage stress.[5] This feature may allow for storage of information in a rewritable manner. For example, bias voltage stress applied to films of poly(*p*-phenylene vinylene) doped with mobile ions, sandwiched between two non-Ohmic contacts, induces the formation of a junction and results in diode-like current−voltage (*I*−*V*) characteristic.[5] After junction formation these cells can emit electroluminescence (light-emitting electrochemical cell, LEEC). The electrical response of these devices to positive and negative bias voltage stress is very similar and both types of stress induce a high conductivity state for probe voltages that have the same parity as the applied bias stress.[5] This latter property makes it difficult to use these structures as memory cells in a passive matrix array because a particular cell in the matrix array will experience reverse bias stress when trying to reduce the conduction level or when trying to raise the conduction level in another cell. This may result

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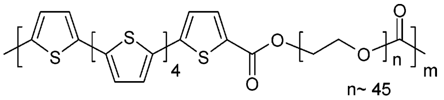

Figure 1. Chemical structure of the 6T-PEO block copolymer.

in creating a diode with the wrong polarity that makes it impossible to read out cells in the matrix array selectively.

Devices with one Ohmic and one Schottky contact feature the required asymmetric response to positive and negative bias stress voltages, but here the induced change in conductivity is mainly in the reverse bias regime.[6] Because modulation of the conduction level in forward bias is required for selective readout of cells in the matrix, these cells are not useful in arrays.

In this paper we report on charge transport in a 6T-PEO block copolymer[7] that contains semiconducting blocks (sexithiophene, 6T) and ion-transporting blocks (poly(ethylene oxide), PEO) alternating along the polymer chain (Figure 1). The use of this alternating block copolymer ensures nanoscopic mixing of the electron- and ion-transporting components, resulting in unique properties. We contact these films on one side with an electrode consisting of a conducting π-conjugated polymer (PEDOT:PSS), doped with an inorganic salt (NaCl) and a plasticizer (ethylene carbonate, EC), and on the other side with a vacuum-deposited Al electrode. In this way a diode configuration is created for which the conductivity in the forward direction can be modulated by application of forward and reverse bias stress. Thus, information can be stored in the cell in a reversible manner. Importantly, negative bias voltage stress does not raise the conduction level under the reverse bias. Therefore, the cell meets an important requirement for passive matrix addressing, illustrating the potential of molecular engineering to design switchable electronic elements.

Experimental Section

The devices were built on a glass substrate with a transparent tin-doped indium oxide (ITO) electrode. An aqueous dispersion of poly(ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) containing NaCl (20 wt % relative to the weight of dry material in the dispersion) and EC (20 wt %) was spin-coated on the ITO to create the bottom electrode. The resulting PEDOT:PSS(NaCl, EC) layer was dried (2 min, 180 °C, thickness 0.07 μm) and a layer of 6T-PEO was deposited by spin-coating from 15 mg/mL tetrahydrofuran (THF) solution (thickness 0.09 μm). The top metal contact was made by vapor deposition of Al at 5×10^{-6} mbar. The active area of the devices was 0.095 cm^2. Devices were stored and measured in an inert atmosphere (O_2, H_2O $\sim$ 1 ppm) using a Keithley 2400 source meter. Cyclic voltammograms were recorded in an inert atmosphere with 0.1 M tetrabutylammonium hexafluorophosphate (TBAPF$_6$) in dichloromethane (DCM) or THF as supporting electrolyte. Redox potentials were measured against an Ag/AgCl reference electrode referenced in situ against Fc/Fc$^+$.

Results and Discussion

The cyclic voltammogram of 6T-PEO reveals that the first oxidation and reduction waves are chemically reversible (Figure 2). The first oxidation potential observed at +0.43 V vs Fc/Fc$^+$ is close to that of α,ω-dihexylsexithiophene (+0.42 V).[8] The first reduction of the 6T-PEO occurs at -2.09 V vs Fc/Fc$^+$, i.e., less negative than that for α,ω-dihexylsexithiophene (-2.23 V). This difference is attributed to the electron-withdrawing effect of the ester moieties in 6T-PEO. The lower part of Figure 2 shows an approximate band level diagram of the devices under study, featuring the HOMO and LUMO levels of the 6T block as determined from the cyclic voltammetry. The lowest barrier for charge carrier injection into 6T-PEO is for holes entering via the PEDOT:PSS electrode. The barrier at the opposite electrode is considerably higher and, therefore, diode-like behavior is expected.

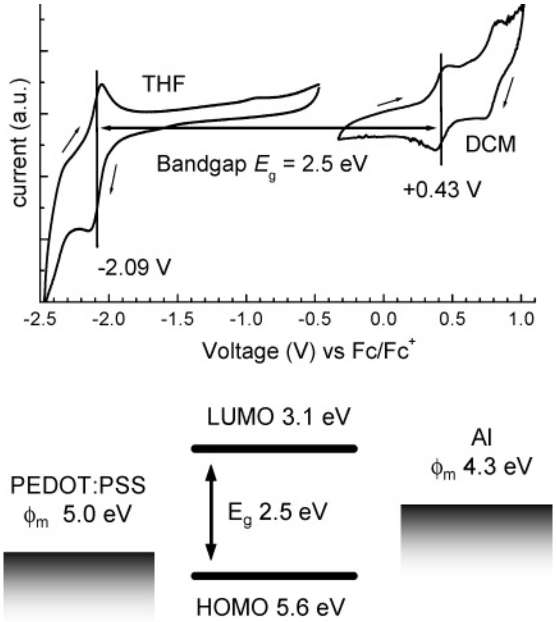

Figure 2. Cyclic voltammetry data for 6T-PEO in tetrahydrofuran (THF) and dichloromethane (DCM). The lower part represents a band level diagram for the devices under study.

The absorption spectrum of 6T-PEO in THF shows the absorption band associated with the optically allowed transition from the ground state (S_0) to the lowest excited singlet state (S_1) with a maximum at 2.8 eV, in agreement with data for sexithiophene (Figure 3).[9] For a solid film of 6T-PEO on glass, the maximum absorption in the UV$-$vis spectrum shifts to higher photon energies (3.2 eV), while the onset occurs at lower energies. These changes are accompanied by a shift of the 6T-PEO photoluminescence to lower photon energies. These spectral changes indicate that the 6T units of 6T-PEO cluster in the solid state and form an H-type aggregate with the 6T moieties stacking in a cofacial or herringbone type fashion with their long axis almost perpendicular to the line joining the centers of the 6T units.[10] Similar spectral features have been observed upon crystal-

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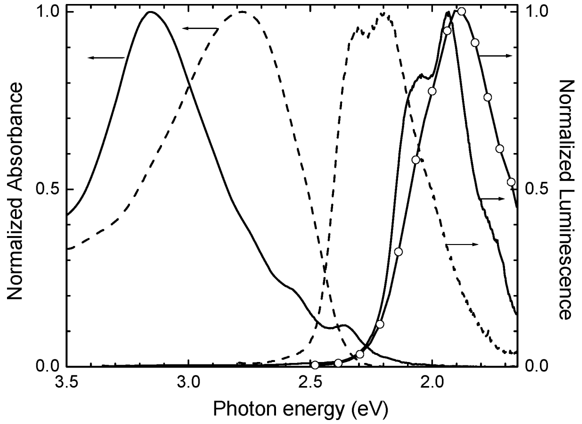

Figure 3. Normalized absorption and photoluminescence (excitation photon energy 3.5 eV) spectra of the 6T-PEO block copolymer as solid film spin-cast from tetrahydrofuran (THF) solution (solid lines) and dissolved in THF (10^{-5} M) (dashed lines). Electroluminescence from a ITO/PEDOT:PSS-(NaCl,EC)/6T-PEO/Al structure (solid line with open circles).

lization of sexithiophene[11] and aggregation of a number of derivatives similar to 6T-PEO.[10]

The current−voltage ($I-V$) characteristic of a ITO/PEDOT:PSS/6T-PEO/Al device without salt or plasticizer added to the PEDOT:PSS reveals clear diode behavior (Figure 4a). At +4 V forward bias (Al electrode negatively biased with respect to the PEDOT:PSS electrode), the current density is approximately 2 orders of magnitude higher than at −4 V bias. Similar alternating block copolymers with oligothiophene blocks shorter than 6 thiophene units[12] did not show diode behavior when tested under identical conditions. The observation of electrical conductivity in 6T-PEO in combination with the insulating properties of the PEO blocks imply the presence of percolating pathways from one electrode to the other via adjacent 6T moieties, enabling transport of charge carriers. This view is supported by the aggregation of the 6T blocks inferred from UV−vis absorption spectroscopy and supports the conclusion that phase separation of the two blocks occurs.

In Figure 4a the scan direction of the $I-V$ characterization is indicated. When scanning back from positive bias toward negative bias, the current changes sign at +2.4 V. In a separate experiment we determined the open-circuit voltage (V_{oc}) displayed by the cell after being biased at +6 V for 60 s (Figure 5a) This voltage amounts to +2.4 V and this is virtually identical to the difference between oxidation and reduction potentials as determined from CV in solution (Figure 2). This correspondence, which has also been observed for LEECs,[5] indicates that V_{oc} originates from Faradaic charging facilitated by a small number of mobile ions originating from, e.g., the PEDOT:PSS layer. Under forward bias, the undoped devices work as the charging of a battery with oxidation of the neutral 6T block taking place at the PEDOT:PSS electrode and reduction of neutral 6T at the Al electrode. After charging for 60 s at +6V, and subsequently short-circuiting the device, electrical current flows spontaneously, decaying with time (Figure 5b). By integration over the time window of observation, we find that 6 × 10^{-5} C/cm^2 flows from the cell. When monitoring the current density while applying a constant bias voltage (Figure 5c), we find that the current density reaches a maximum at ~6 s after switching on the bias voltage. This indicates that the charging has reached completion, probably because the source of mobile ions is exhausted.

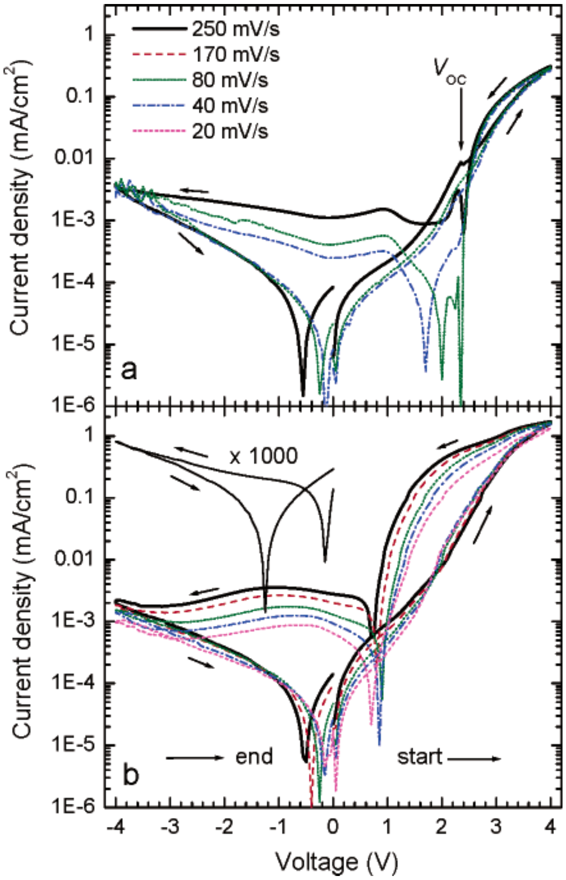

Figure 4. Current−voltage characteristics of ITO/PEDOT:PSS/6T-PEO/Al devices at different scan speeds. (a) No salt or plasticizer added. (b) With NaCl and EC added to PEDOT:PSS. The trace in the upper left corner of panel (b) pertains to a scan of only the reverse bias voltages and was taken directly after a 1 min period of −4 V bias stress (scan speed 170 mV/s).

ITO/PEDOT:PSS(NaCl,EC)/6T-PEO/Al devices to which salt and plasticizer have been added deliberately show current densities under forward bias that are approximately 1 order of magnitude higher than those for the undoped devices (Figure 4b). A large hysteresis is now observed in the $I-V$ characteristics, which results from the interdependence of the field-induced transport of ions toward the electrodes and electrochemical redox reactions of the 6T block of the polymer. This combination of processes is well-known from research on LEECs.[5] The ion accumulation and electrochemical doping at the electrodes strongly reduce the barrier for charge injection. This combination of processes results in a hysteresis with an anticlockwise direction. In contrast, the hysteresis in the cyclic voltammetry scan has a clockwise direction when the absolute value of the current is plotted (Figure 2). This underlines that the hysteresis observed for the diodes does not originate only from reversible redox reactions at the electrodes but results from junction formation.

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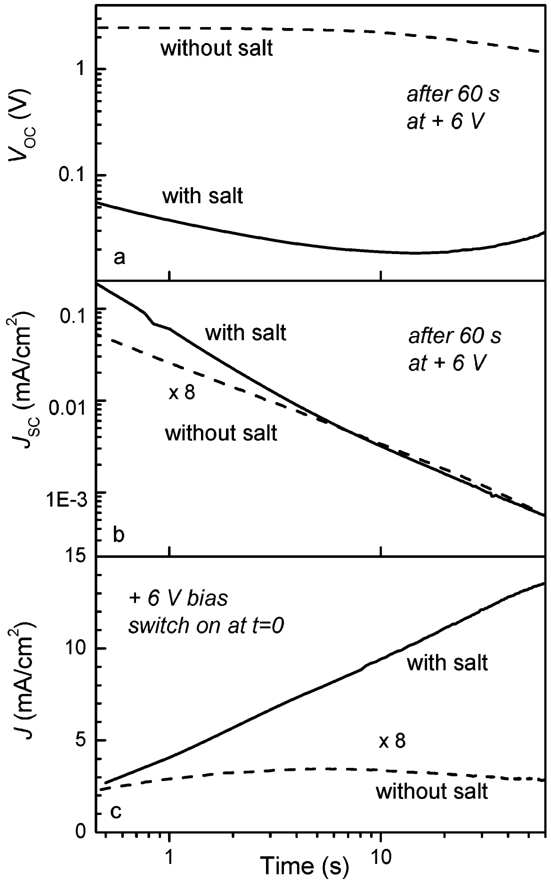

Figure 5. (a) Time dependence of the open-circuit voltage (V_{OC}) measured directly after termination of a bias voltage stress (+6 V, 60 s) for a PEDOT:PSS/6T-PEO/Al diode (dashed line) and of a PEDOT:PSS(NaCl,EC)/6T-PEO/Al diode (solid line). (b). Short-circuit current (J_{SC}) after bias stress (+6 V, 60 s). (c) Current density (J) under +6 V bias stress started at $t = 0$.

The open-circuit voltage for the doped devices, measured directly after 60 s at +6 V bias stress, drops below 0.1 V within 1 s after termination of the stress voltage (Figure 5a). While the devices without added salt show a saturation of the current density at about 10 s after the turn on of the bias voltage, the devices with added salt do not show a clear saturation in the period of measurement (Figure 5c).

Interestingly, devices with added salt retain their diode characteristic. The current density under negative bias is 3 orders of magnitude lower than that in the forward (Figure 4b). The application of reverse bias voltages does not raise the conduction level in the reverse bias regime. To confirm the absence of any ion-induced electrode processes under reverse bias, we have biased the devices for 60 s at −4 V and performed an $I-V$ scan of the negative bias region. The result shown in the upper left corner of Figure 4b reveals no enhancement of the current density at negative bias. This indicates that migration of anions toward the interface with the Al electrode, leading to a buildup of negative space charge and allowing for facile hole injection through the Al electrode, does not take place. In principle, ion migration induced by an electrical field may not be fully reversible. However, in the course of our investigations, we found no obvious signs of fatigue and the cells can be cycled at least 15 times without any significant changes in the $I-V$ characteristic. (See Supporting Information, Figure S1.)

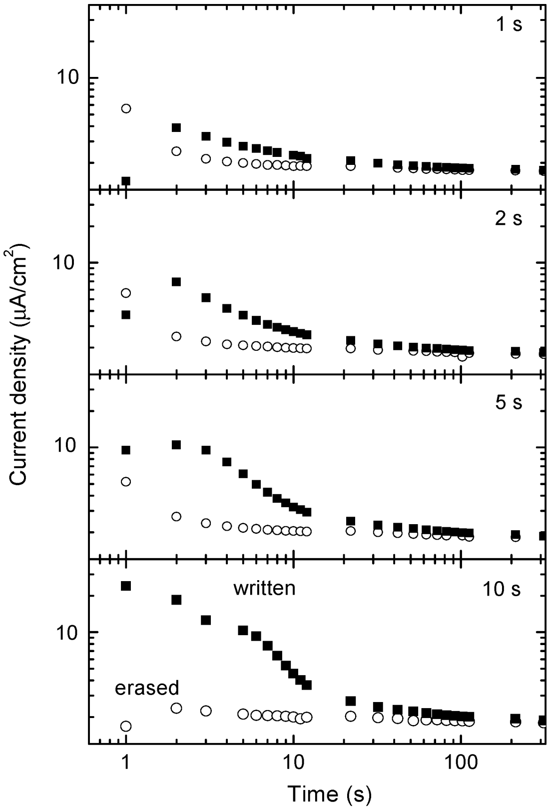

Figure 6. Time dependence of the current density of the ITO/ PEDOT: PSS(NaCl,EC)/6T-PEO/Al at $V = +1.5$ V after a +4 V write pulse (■) and after a −4 V erase pulse (○). The length of the write and erase pulses is systematically varied from 1, 2, 5, and 10 s. The read pulses have a temporal width of ~40 ms and between the read pulses the devices are kept at 0 V bias.

The response of the devices to pulses of positive and negative bias voltages has been analyzed in more detail. A bias voltage of +4 V was applied for 1 s and subsequently the conductivity of the diode was probed by applying multiple short (~ 40 ms) "read" pulses of +1.5 V and monitoring the current density (Figure 6, upper panel, solid squares). Between the read pulses, the device was kept short-circuited. Two transient components to the current density under the read voltage may be expected. The first is a Faradaic discharge current which flows in a direction opposite to the steady-state current under +1.5 V bias when the device has been stressed at +4 V. The second transient contribution results from the enhanced injection of charge carriers at +1.5 V after modification of the barriers for injection by the motion of ions and the associated redox reactions resulting from the +4 V stress. This excess current flows in the same directions as the steady-state current at +1.5 V bias. We find that at times $t < 1$ s after the write pulse, the Faradaic discharge current dominates while for times $t > 1$ s, the current density is enhanced due to the junction formation. When the +4 V bias is applied for a longer time interval (2, 5, or 10 s), the importance of the discharge current ($t < 1$ s) is reduced while the excess current ($t > 1$ s) due to junction formation increases. In Figure 6 we also show the response to a −4 V bias voltage pulses (open circles). Here, the discharge current has the same direction as the steady-state current under +1.5 V bias. Therefore, the enhanced current densities in the first second

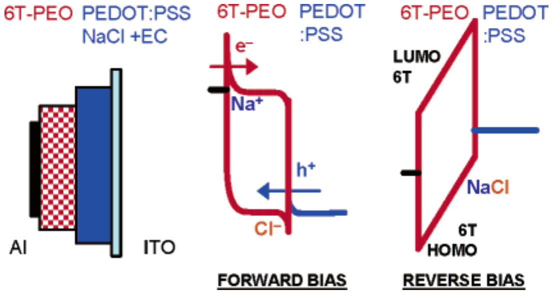

Figure 7. Schematic layout of the ITO/PEDOT:PSS(NaCl,EC)/6T-PEO/Al device (left) and a band level diagram for the diode under forward bias stress (middle) and reverse bias stress (right). The injection of negative (e^-) and positive (h^+) charge carriers into the semiconducting block of the copolymers is indicated.

after the −4 V pulse as observed for the 1, 2, and 5 s voltage pulses may be ascribed to the discharge current. Consistent with the absence of junction formation under reverse bias as inferred from the $I-V$ scans, we find no significant reduction of the current density at the read voltage due to induction of a junction with opposite polarity by applying −4 V bias stress.

This asymmetric response of the diodes and the excess current density associated with the hysteresis allows us to temporarily store information in the cells in a reversible manner. The cells can be "written" at +4 V, "erased" at −4 V, and "read" in forward bias at +1.5 V. As can be seen from Figure 6, the excess current density allows one to distinguish a low-conductivity state and a high-conductivity state in the time window between 1 and 10 s when the write and erase pulses are ≥5 s. For delays larger than 10 s after the write/erase pulse, the current densities for the written and erased states quickly converge. Thus, the retention time of the information (10 s) is at present rather short. To improve on this, a block copolymer with a very steep dependence of the ion mobility on electric field is needed.

As mentioned above, the hysteresis in forward bias can be rationalized in terms of electrochemical doping at the electrodes induced by ion transport.[5] The effect of ion accumulation and electrochemical doping on the barriers for charge injection are illustrated schematically in Figure 7. Under application of forward bias stress, electrochemical doping of 6T occurs at both electrodes. Near the Al, reduction of the 6T is facilitated by migration of sodium ions toward the metal electrode (n-type doping). Oxidation of 6T occurs near the interface with the PEDOT:PSS electrode and is associated with migration of the chloride ions in the vicinity of the interface (p-type doping). This results in formation of a p−n junction.[13,14] This mechanism is supported by the temperature dependence of the $I-V$ characteristic. At low temperature, where motion of ions is inhibited, the hysteresis is strongly reduced. Conversely, at high temperature, the mobility of ions is higher and relaxation to the thermodynamically favored state is faster. Hence, the hysteresis is suppressed as well (Figures S2 and S3, Supporting Information). As expected, the hysteresis remains when taking top metal contacts with different work functions (e.g., Au or Ca, Figure S4).

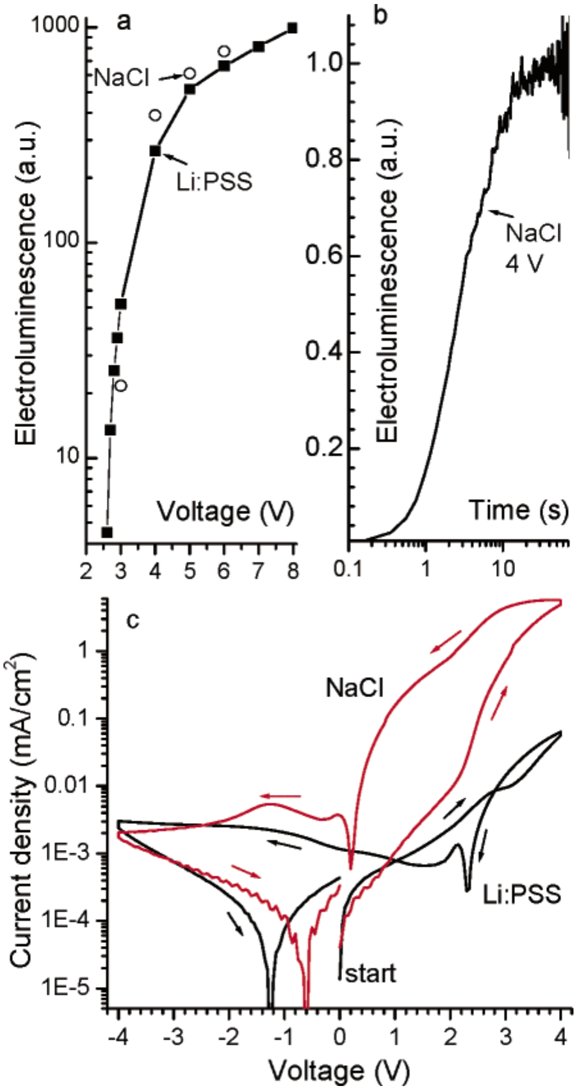

Figure 8. (a) Electroluminescence intensity as a function of applied voltage for ITO/PEDOT:PSS/6T-PEO/Al diodes doped with EC and either NaCl (open circles) or Li:PSS (20 wt %, solid squares). (b) Time dependence of the electroluminescence intensity for the diode with NaCl. At time $t = 0$ s a constant bias voltage of +4 V was switched on. (c) $I-V$ characteristics of the diodes with Li:PSS or NaCl. Scan speed 170 mV/s, room temperature.

To account for the low open-circuit voltage after bias stress as observed for the diodes with salt, we argue that a p−n junction is formed with a narrow depletion zone between the p and n doped regions. Such narrow widths are expected when extensive electrochemical doping occurs.[15] Because of the narrow depletion width, mobile electrons and holes on the 6T segments can recombine after diffusional motion, reducing any open-circuit voltage quickly after termination of the bias voltage.

The observation of electroluminescence (EL, Figure 3) allows us to analyze the junction formation in further detail. EL is observed for devices when the PEDOT:PSS layer is doped with either NaCl or Li:PSS. The EL shows an onset at +2.5 V (Figure 8a), which is delayed in time with respect to the start of the bias voltage (Figure 8b). To generate EL, electrons must be injected via the Al electrode, a process

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that is assisted by accumulation of small positive ions (Na^+, Li^+) near the Al electrode and after being transported through the block copolymer. The delayed onset of the EL with respect to the application of the bias voltage can then be interpreted in terms of the time needed for the transport of ion and electrochemical doping processes.

Interestingly, for the devices with Li:PSS added, the hysteresis in the $I-V$ characteristic is strongly reduced in comparison with devices doped with NaCl (see Figure 8c). Because of its bulkiness, the PSS polyanion will hardly migrate. The fact that EL is observed upon doping with Li:PSS in combination with the absence of a considerable hysteresis in the forward bias regime indicates that the hysteresis observed with NaCl doping results from migration of the Cl^- anions to the vicinity of the PEDOT:PSS/6T-PEO interface, resulting in an effective lowering of the barrier for hole injection. From the absence of enhanced current under reverse bias stress, we infer that the Cl^- anions are not transported through the block copolymer to the interface with the Al electrode. Had the chloride ions come into close proximity of the Al electrode, electrochemistry would be expected, resulting in an Ohmic contact for hole injection. In this interpretation the asymmetry of the diode in its response toward positive and negative bias stress results from selective transport of ions.

Conclusion

In summary, we have shown that block copolymers with a semiconducting and ion-transporting block can be used to construct diodes displaying resistive switching in the current under forward bias without any appreciable modulation of the conductivity in the reverse bias regime. This type of behavior is required for integration of memory cells in passive matrix arrays. The asymmetry in the response is attributed to selectivity in the transport of ions by the block copolymer. Molecular engineering of block copolymers combine charge and ion transporting moieties; therefore, it seems to be a promising tool to design switchable electronic elements.

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Supporting Information Available: Requirements to rewritable passive matrix addressing diodes exhibiting resistive switching behavior. Figures S1−S4. This material is available free of charge via the Internet at http://pubs.acs.org.

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