

Available online at www.sciencedirect.com



Polymer 46 (2005) 7771-7778

polymer

www.elsevier.com/locate/polymer

Synthesis and characterization of conducting interpenetrating polymer networks for new actuators

Cédric Plesse, Frédéric Vidal, Hyacinthe Randriamahazaka, Dominique Teyssié, Claude Chevrot*

Laboratoire de Physicochimie des Polymères et des Interfaces (LPPI), Université de Cergy-Pontoise, 5 Mail Gay-Lussac, F-95031 Cergy-Pontoise Cedex, France

> Received 18 October 2004; received in revised form 18 February 2005; accepted 15 March 2005 Available online 11 July 2005

Abstract

The synthesis of conducting interpenetrating polymer networks (IPNs) for actuator applications is described. The IPNs are synthesized from poly(ethylene oxide) and polybutadiene networks in which the conducting polymer (poly(3,4-ethylenedioxythiophene)), PEDOT, is gradually dispersed, i.e. its content decreases from the outside towards the centre of the film. The conducting IPN morphology was investigated by electrical resistance measurements and DMA. For actuation experiments, aqueous or organic electrolyte solutions were used but the best results were obtained with a room temperature ionic liquid, 1-ethyl-3-methylimidazolium bis-(trifluoromethylsulfonyl)imide (EMITFSI).

© 2005 Elsevier Ltd. All rights reserved.

Keywords: Interpenetrating polymer networks; Actuators; Synthesis

1. Introduction

Among electronic conducting polymers (CP), poly(3,4ethylenedioxythiophene) (PEDOT) has received much attention recently because of its particularly high stability in the doped state and the reversibility of the doping process [1,2]. However, PEDOT as many conducting polymers is insoluble in common solvents and the conventional methods of polymer processing cannot be used. Extensive studies have been carried out in order to overcome the poor processability of such compounds [3]. Improvements are expected by combining conducting and thermoplastic or elastomer insulating polymers in which the conducting polymer furnishes the required conducting properties, while the insulating polymer matrix improves the mechanical properties. One possibly promising solution is the combination of a conducting and an insulating polymer into an interpenetrating polymer networks (IPNs) super structure. Indeed, the presence of cross-linked polymers in these

architectures ensures both dimensional stability and the requested mechanical properties [4]. Also the entanglements between the cross-linked polymers increase the miscibility with the CP compared to usual blends. In most reported cases, the measured volume percolation threshold for conductivity (generally below than 5%) is lower than that which is observed in statistical blends (e.g. 16 vol%) [5–7].

Conducting polymers have also attracted attention because of dimensional change properties due to expulsion/ inclusion of ions during doping/dedoping processes which lead to interesting potential applications in the design of actuators for robotics, prosthetics, microvalves [8-13]... Air working actuators are generally built in a configuration, where the internal layer is a solid polymer electrolyte (SPE) sandwiched between two CP layers (CP//SPE//CP) and the relative differential expansion between conducting polymer layers results in bending [14]. However, when a conductive polymer film is deposited onto a flexible polymer film sputtered with a thin layer of metal, a delamination process which limits the actuator's lifetime is almost always observed [15]. As we have reported previously [16,17] this problem can be overcome if the actuator is designed as a conducting interpenetrating polymer network (IPN).

^{*} Corresponding author.

^{0032-3861/}\$ - see front matter © 2005 Elsevier Ltd. All rights reserved. doi:10.1016/j.polymer.2005.03.103

In this study, the synthesis of conducting IPNs leading to actuator applications is described. The CP has been prepared from oxidative polymerization of the corresponding heterocyclic monomer inside the IPN matrix. This IPN matrix is made from poly(ethylene oxide) (PEO) and polybutadiene (PB) networks chosen as an interesting solution to the design of solid polymer electrolytes (SPEs). The PEO network acts here as the SPE partner, whereas the PB network ensures good mechanical properties preventing crack formations during bending deformations. The PB network is formed by polyaddition of the OH functions of dihydroxytelechelic polybutadiene (HTPB) with a NCO-crosslinkers (Desmodur® N3300) and the PEO network is obtained by radical copolymerization of poly(ethylene glycol) dimethacrylate and methoxy poly(ethylene glycol) methacrylate. Conducting IPNs are synthesized from PEDOT and the above mentioned SPEs. The synthetic pathway leads to a gradual dispersion of the electronic conducting polymer through the thickness of the network, i.e. the quantity of PEDOT decreases from the outside towards the centre of the film. Finally application to actuators working in open air for duration over one month is demonstrated.

2. Experimental

2.1. Materials

Methoxy poly(ethylene glycol) methacrylate (PEGM, $M_{\rm w} = 300 \text{ g mol}^{-1}$), poly(ethylene glycol) dimethacrylate (PEGDM, $M_{\rm w} = 875 \text{ g mol}^{-1}$), polybutadiene— α, ω dihydroxy functionalized (HTPB, $M_{\rm w} = 2800 \text{ g mol}^{-1}$), and dibutyltin dilaurate (DBTDL 95%) were obtained from Aldrich and used without further purification. 3,4-ethylenedioxythiophene (EDOT) (Bayer) was distilled under reduced pressure prior to use. Acetonitrile (HPLC grade from Acros), dichloromethane (Carlo-Erba), propylene carbonate (HPLC grade from Carlo-Erba) methanol (Carlo-Erba), anhydrous iron III chloride (Acros), lithium perchlorate (Aldrich), dicyclohexylperoxidicarbonate (DCPD) (Groupe Arnaud) and Desmodur[®] N3300 (21.8 \times 10^{-3} mol of NCO for 1 g of Desmodur) (Bayer) were used as received. The synthesis of the room temperature ionic liquid (RTIL), 1-ethyl-3-methylimidazolium bis-(trifluoromethylsulfonyl)-imide (EMITFSI), was carried out according to the procedure described by Grätzel et al. [18] for subsequent use as an electrolyte in the conducting IPN.

2.2. Preparation of poly(ethylene oxide)/polybutadiene IPN SPEs (PB/PEO IPN)

DCPD is used for methacrylate function radical initiation of PEGDM and PEGM. Desmodur[®] N3300 is the HTPB crosslinker and DBTDL the polyaddition catalyst between NCO and OH functions. The mixture

(PEGDM/PEGM/HTPB: 0.2/0.6/0.2 weight proportions) was poured into a glass mould to yield a 250 µm thick film. The mould was heated at 50 °C for 3 h and postcured for 1 h at 80 °C. The general procedure synthesis and the DMA characterization of the PEO/PB IPN are described elsewhere [16].

2.3. Preparation of conducting polymer based (semi) IPN

The SPE films were soaked into pure EDOT for given lengths of time and their surface was wiped off with filter paper afterwards. The swollen films were then immersed into a FeCl₃ aqueous solution (1.5 mol L^{-1}) for given lengths of time. The film is then washed several times with methanol until the color disappears completely, i.e. excess of FeCl₃ is removed. The film surface is then wiped off with filter paper. The conducting material is dried at 60 °C under vacuum for 24 h.

2.4. Characterizations

In order to examine the swelling capacity, PEO/PB IPN films (approximate dimensions $10 \times 20 \times 0.250 \text{ mm}^3$) with accurately known weights were immersed (either in pure EDOT or EMITFSI) at room temperature. The films were removed from each liquid at different times and carefully wiped. The swelling ratio in the samples is defined as:

$$SR(\%) = \frac{(W_s - W_0)}{W_0} \times 100$$
(1)

where W_0 and W_s are the weights of SPE samples before and after swelling.

In order to examine whether the SPE synthetic pathway leads to an adequately tuned material, salt uptake (LiCl, CaCl₂, BaCl₂, tBuNClO₄ and LiClO₄) measurements were performed. IPNs were swollen with aqueous salt solutions $(10^{-1} \text{ mol L}^{-1})$ for different times. Then samples were dried overnight at 50 °C under vacuum and the salt uptake was calculated.

Dynamic mechanical analysis measurements are carried out on single network, IPN or semi-IPN samples (typically $L \times w \times t = 15 \times 8 \times 0.5 \text{ (mm^3)}$) with a Q800 model (TA Instruments) operating in tension mode (strain between 0.05 and 0.07%, pretension 10^{-2} N). Experiments were performed at 1 Hz frequency and with a heating rate of 2 °C/min from -100 to +50 °C. The set up provides the storage and loss moduli (E' and E'') and the damping parameter or loss factor (tan δ) defined as the ratio tan $\delta = E''/E'$.

The two terminal-technique with gold pressure contact was used for the room temperature electrical resistance measurements on PEO/PB/PEDOT IPNs. Electrical resistance measurements were performed along the surface (R_s) and across the thickness or bulk (R_b) with a KEITHLEY 197 autoranging microvolt DMM. For the measurement of R_s , the interspacing of electrodes (area of 1.8 mm^2) was 1 cm. The bulk conductivity was measured after cutting the film edges in order to suppress the electrical conductivity contribution through the edges along the surface.

2.5. Actuator capacity

After the edges were cut out, the conducting semi-IPNs (approximate dimensions $10 \times 30 \times 0.250 \text{ mm}^3$) were immersed in a chosen electrolyte solution for a given time at room temperature. The conducting IPNs were then maintained horizontal with steel clamps in order to ensure electrical contact with the VMP Bio-logic (Science Instrument) potentiostat. The bending response was obtained by applying a voltage between ± 2 and ± 5 V.

A data acquisition system was used to allow the actuator bending deformation. Using a CCD ToUcam USB camera, the actuator position was characterized by calculating the curvature ρ (cm⁻¹) as described elsewhere [19].

Actuator capacity was tested at room temperature and at -30 and -20 °C using a climatic test chamber Binder MK53.

3. Results and discussion

3.1. Polybutadiene/poly(ethylene oxide) IPN SPEs (PB/PEO IPN)

3.1.1. Synthesis

In a previous study, PEO/polycarbonate IPNs were prepared as our first attempt to design conducting polymer based actuators [11,20] However, for this particular chemical composition, bending deformations lead to crack formations at room temperature since deformations induce stress inside the material. To prevent cracking, polybutadiene (PB) was used instead of polycarbonate [16]. This elastomer was shown to be able to absorb large deformations.

Therefore, SPE IPNs based on PEO and PB with different relative weight proportion were prepared, mixing all components of the two partner networks from the start. The PEO network is obtained from methacrylate end group polymerization/cross-linking in the presence of DCPD as initiator. The PB network is obtained through cross-linking reactions between dihydroxy telechelic polybutadiene (HTPB) and Desmodur[®] in the presence of DBTDL as catalyst. The synthesis of IPNs requires a minimum of toluene as solvent since HTPB and PEGDM/PEGM are not miscible. The mixture is heated at 50 °C for 3 h, leading to the formation of the IPN. The morphology of the resulting IPN has been described elsewhere [17] and typical dual phase morphology is observed for composition ranging from 90 to 50 wt% PEO in the mixture. For PEO wt% less than 40% composition, isolated PEO domains appear and the PB phase is continuous in space.

3.1.2. Swelling studies of PEO/PB IPNs

3.1.2.1. EDOT swelling study. According to the procedure described in Section 2, the mass quantity of EDOT loaded in the SPEs can be controlled by the duration of the swelling in pure EDOT. This mass quantity measures the amount of EDOT that is trapped within the SPE matrix and proved perfectly reproducible. The swelling study of the single PEO network could not be performed since a fragmentation of the sample occurs after 2 min immersion. For the single PB network, a plateau value around 60 wt% EDOT is obtained after 60 min swelling as depicted on Fig. 1. The EDOT swelling kinetics of various PEO/PB IPNs show that the maximum EDOT uptake increases from 100 to 350% for IPNs containing 10 to 90 wt% PEO, respectively. Furthermore the swelling rate increases with increasing weight proportion of PEO. This high affinity may reflect a specific interaction between the ethylenedioxy function of EDOT and the ethylene oxide units in the PEO chains. It is also worthwhile to note that for IPNs containing the highest weight proportion of PEO, i.e. 80 and 90 wt%, no destruction of samples is observed contrary to what happened with single PEO networks. This result can be taken as an evidence of the synergy of properties in the IPNs.

In the forthcoming, only results concerning IPNs with a relative weight proportion PEO/PB 80/20 will be discussed since these particular materials were used as a matrix for actuator applications.

3.1.2.2. EMITFSI swelling study. The room temperature ionic liquid ethyl-3-methylimidazolium bis-(trifluoromethylsulfonyl)imide (EMITFSI) can be used as electrolyte for actuator application. Therefore, the mass quantity of EMITFSI, which can be absorbed by the IPN, was measured. Fig. 2 shows the EMITFSI uptake kinetics in the PEO/PB (80/20) IPN as well as in single PEO and PB networks. First of all, EMITFSI does not enter into the PB matrix at all, even if this ionic liquid is usually considered as a hydrophobic salt. In more polar matrix such as the PEO



Fig. 1. EDOT swelling kinetics for (a) single PB network, and for PEO/PB IPNs (x/1-x). x = (b) 90 wt%, (c) 80 wt%, (d) 70 wt%, (e) 60 wt%, (f) 50 wt%, (g) 40 wt%, (h) 30 wt%, (i) 20 wt%, (j) 10 wt%.



Fig. 2. EMITFSI swelling kinetics for (\blacktriangle) single PEO network, (\diamondsuit) single PB network, and for (\Box) PEO/PB IPN (80/20) wt/wt.

single network or the IPN, the EMITFSI uptake is quite high, and reaches 200 wt%, which indicates a high affinity between EMITFSI and PEO. It should be noticed that a partial destruction of the single PEO network is observed but not in the case of the IPN. Again, the synergy of properties brought by the combination of PEO into an IPN with PB is demonstrated.

3.1.3. DMA characterization

The typical viscoelastic behaviour of the PEO/PB (80/20) IPN was studied by DMA. Fig. 3 shows the loss tangent (tan δ) as a function of temperature. Only one relaxation is observed (-45 °C) which is due to the close relaxation temperatures of the two single networks (-63 and -45 °C for single PB and PEO networks, respectively).

DMA characterization of the EMITFSI swollen PEO/PB IPN has been also carried out. Fig. 3 shows also the alpha relaxation of EMITFSI swollen PEO/PB IPN. In this case the alpha relaxation is 15 °C lower than that of the unswollen IPN demonstrating the plasticizing effect of EMITFSI. This result also indicates that EMITFSI is not able to crystallize inside the matrix at -60 °C, temperature



Fig. 3. Dynamic thermo-mechanical spectroscopy. Tan δ of (PEO/PB) IPN (80/20), without (- - -) and with (—) EMITFSI.

which is very low compared to that of the melting point of EMITFSI [18] (-3 °C).

3.2. Conducting IPNs (PEO/PB/PEDOT IPN)

3.2.1. Synthesis

Conducting IPNs were prepared by dipping the EDOT swollen IPNs films into an iron III trichloride (FeCl₃) solution. Different solvents were used for FeCl₃ (chloroform, acetonitrile...) and it was observed that the locus of EDOT polymerization, and thus the conductivity of the resulting IPN depends on the nature of solvent. It has been observed that the exit rate of EDOT from the film relative to the entry rate of FeCl₃ in the matrix seems to play a major role [11]. The best results are obtained if water is chosen as the solvent for the oxidative polymerization of EDOT. Indeed due to the low EDOT solubility in water, i.e. 1.5×10^{-2} mol L⁻¹, the polymerization rate and the diffusion of EDOT out of the film into the aqueous solution is slow and allows the penetration of FeCl₃ in the matrix before a significant amount of EDOT leaves the matrix.

After the general experimental conditions (concentration of reactants, solvents...) for the preparation of conducting materials were established, two different pathways were investigated which both lead to conducting IPNs. In the first procedure IPNs with a 300 wt% EDOT content are dipped for different lengths of time into a FeCl₃/water solution $(1.5 \text{ mol } \text{L}^{-1})$. Alternatively the second procedure consists to dip swollen IPNs with different EDOT contents, for 16 h into a FeCl₃/water solution (1.5 mol L^{-1}). Sulfur elemental analyses were performed on all samples. Fig. 4 shows the PEDOT final content in the IPNs measured on the samples prepared through the first procedure, i.e. when the EDOT polymerization is carried out from a 300 wt% EDOT content. The PEDOT content in the IPN increases with the chosen length of the polymerization time (seven samples were prepared with polymerization duration from 2 to 50 h). A maximum of 10% polymerization yield inside the matrix is observed. Indeed a 300 wt% EDOT loaded IPN leads to a maximum of 30 wt% of PEDOT inside the film after 24 h of



Fig. 4. PEDOT weight content in the conducting IPN as a function the EDOT polymerization length time. FeCl₃/water solution: $1.5 \text{ mol } \text{L}^{-1}$. EDOT loaded in the PEO/PB IPN 80/20: 300 wt%.

polymerization. This result means that the main locus of EDOT polymerization is outside the film, either in the water or at the film surface.

Alternatively (Fig. 5), for a constant time of EDOT polymerization (16 h) an increase of the initial weight proportion of EDOT loaded in the IPN (from 70 to 300 wt%) leads to an increase of the final PEDOT content. This second procedure does not lead to an increase in the polymerization yield, which remains again here below 10%.

3.2.2. Characterization

For all samples, the electrical resistance was measured along the surface (R_s) and across the thickness or bulk (R_b) of the film. The $R_{\rm b}/R_{\rm s}$ ratios are plotted on Figs. 6 and 7 as a function of the weight proportion of PEDOT according to the first and second synthesis procedure, respectively. Each $R_{\rm b}/R_{\rm s}$ curve exhibits a maximum, which shows that the bulk resistances are at least 100 times higher than the resistance measured along the surface. Furthermore, the electrical resistance R_s reaches a plateau value around 10–30 Ω for PEDOT contents higher than 10 wt%. This result implies that there is an inhomogeneous distribution of PEDOT across the sample thickness, i.e. that the PEDOT concentration decreases from the outside towards the centre of the IPN sample, leading to a very poor connectivity of PEDOT inside the bulk of the matrix. This behaviour can be taken advantage of especially bearing in mind the fact the preparation of IPNs with a gradual dispersion of the conducting polymer throughout the thickness would be an asset for actuator applications. Furthermore this gradual dispersion has been confirmed by energy dispersion spectrometer (EDS) [17]. A 10 µm depth from each side of the sample has been observed.

As shown on Fig. 8, the EMITFSI uptake kinetics decrease drastically in the case of conducting IPN compared to the PEDOT depleted PEO/PB IPN. However, the maximum uptakes for both IPNs are of the same order of magnitude, i.e. 200 wt%. Thus the conducting polymer



Fig. 5. PEDOT weight content in the conducting IPN as a function the EDOT weight proportion loaded in the PEO/PB IPN 80/20. EDOT polymerization time: 16 h.



Fig. 6. Variation of R_b/R_s ratio as a function of the weight percent of PEDOT. First synthesis procedure of conducting IPNs: 300 wt% EDOT loaded in the PEO/PB IPN are dipped for different lengths of time into a FeCl₃/water solution (1.5 mol L⁻¹).

10 μ m layer forms a shield that limits the inwards diffusion rate of EMITFSI.

3.2.3. DMA characterization

The influence of PEDOT content in the IPNs on the viscoelastic properties of the materials is shown in Fig. 9(a) and (b). As the weight proportion of PEDOT increases the rubber plateau value increases and conversely the damping properties decrease. Thus at room temperature the stiffness of samples increases with the PEDOT content. It is noteworthy that the temperatures of the main relaxation (correlated with the glass transition temperature) hardly shift with increasing PEDOT content.

The thermo-mechanical properties of conducting IPNs were also characterized in the presence of EMITFSI. As already observed for PEO/PB IPN (Fig. 3), the ionic liquid also behaves as a plasticizing agent in the conducting IPN. The main relaxation of the conducting IPN is 20 °C lower in the presence of EMITFSI than in its absence (Fig. 10).

3.2.4. Actuator capacity

First of all, only the second synthesis procedure was chosen for the actuator preparation, since this procedure is



Fig. 7. Variation of R_b/R_s ratio as a function of the weight percent of PEDOT. Second synthesis procedure of conducting IPNs: swollen PEO/PB IPNs with different EDOT contents are dipped for 16 h into a FeCl₃/water solution (1.5 mol L⁻¹).



Fig. 8. EMITFSI weight content in (\triangle) the PEO/PB IPN and (\blacksquare) in the PEO/PB/PEDOT IPN as a function of time. PEDOT mean content in the IPN: 7 wt%.

less EDOT consuming (swollen PEO/PB IPNs with different EDOT content are dipped for 16 h into a FeCl₃/ water solution). In these synthesis conditions, the R_b/R_s curve (Fig. 7) shows a maximum for a PEDOT weight content around 7–10 wt%, which corresponds to the steepest PEDOT gradient in the conducting IPNs. Therefore, a series of conducting IPNs were prepared according to this weight proportion for actuator applications.

Before actuation experiments, conducting IPNs were dipped in different salt/solvent combination solutions $(10^{-1} \text{ mol } \text{L}^{-1})$. The mass quantity of electrolyte in the IPN was controlled by the time duration of immersion in the salt/water solution. After salt uptake, a strip of conducting IPN $(10 \times 6 \times$ $0.25 \text{ mm}^3)$ was subjected to a voltage of $\pm 2 \text{ V}$ applied between the two sides of the actuator film. Fig. 11 shows the actuator curvatures as a function of time in open air and demonstrates that these actuators can be operated with different salts and solvents. The best result is obtained with



Fig. 10. Dynamic thermo-mechanical spectroscopy. Storage modulus (–) and tan δ (– – – –) versus temperature of conducting IPNs, (grey): EMITFSI non-swollen sample, (black): 200 wt% swollen sample. PEDOT weight content in the conducting IPN: 7 wt%.

the LiClO₄/propylene carbonate (PC) electrolyte solution which allows the actuator to reach a curvature as high as 1 cm^{-1} in less than 4 s. On the other hand the actuator capacity is greatly affected by the nature of the solvent, since using acetonitrile instead of PC leads to a less efficient actuator: indeed, only a 0.35 cm^{-1} curvature is observed after 50 s. For all tested systems, the conducting polymer (CP) volume increases during the oxidation step and decreases during the reduction step. Many investigations were carried out on the mechanism of the volume change in conducting polymers during the redox process [15]; the mechanism is governed by the incorporation or expulsion of ions and solvents of the CP layer. The size of the anion used in the electrochemical device has a strong influence on the mechanism of charge compensation, therefore, on the volume variation during the redox process. In presence of small anions, anions and solvent molecules motions are involved in the redox process. In this



Fig. 9. Dynamic thermo-mechanical spectroscopy. Storage modulus (a) and tan δ (b) versus temperature of conducting (PEO/PB) (80/20) IPNs. Influence of the weight content of PEDOT: 0, 2.4, 7.2, 12.2 wt%.



Fig. 11. Time dependence of the curvature using as electrolyte/solvent: (a) $LiClO_4/PC$ (25 wt%), (b) $BaCl_2/H_2O$ (6 wt%), (c) $LiCl/H_2O$ (8 wt%), (d) $CaCl_2/H_2O$ (4 wt%), (e) $tBuNClO_4/acetonitrile$ (15 wt%), (f) $LiClO_4/acetonitrile$ (7 wt%). Applied voltage: 2 V.

case, the conducting polymer layer shrinks during the reduction reaction. According to the results obtained above, an anion motion mechanism is postulated here.

Fig. 12 shows that the initial rate of bending increases with the applied voltage [21]. Nevertheless, for applied voltage higher than 2 V, a degradation of the conducting polymer must occur since no bending deformations are observed in these conditions except if the cycling frequency is higher than 1 Hz. For instance under a 8 Hz cycling frequency, the LiClO₄/propylene carbonate swollen actuator can be cycled over 8×10^4 times (3 h) at a voltage up to 3 V before the bending deformation stops due to PC evaporation. However, after being immersed for a few hours in LiCLO₄/PC solution the actuator recovers its working capacity.

In order to prevent solvent evaporation, the actuator was encapsulated into a thin layer of polyisobutene (using the spray coating technique). However, the encapsulation technique only slows down the evaporation but does not prevent it totally since the actuator looses its bending capacity after 24 h. Presently, the best way to avoid the evaporation process is to use a Room Temperature Ionic



Fig. 12. Time dependence of the curvature using as electrolyte $LiClO_4/PC$. Applied voltage: (\diamond) 2 V, (\blacksquare) 5 V, (\triangle) 10 V.

Liquid (RTIL) as electrolyte. Indeed RTILs are non-volatile compounds and have been successfully used for electromechanical actuators [22–24]. In the presence of ethyl-3methylimidazolium bis-(trifluoromethylsulfonyl)imide (EMImTFSI) as RTIL, the actuator could be operated in open air over one month without drying as described elsewhere [16]. Furthermore, the actuator can be stored over a period of 6 months at least without any precaution and be operated again immediately.

According to the results from the DMA characterization, EMITFSI is able to act as an electrolyte even at low temperature. Actuation experiments were carried out at -30 and -20 °C and the time to display a 60° angular deformation upon 3 V was recorded. At -30 °C, the actuator bends within 10 min, which is 60 times slower than at 30 °C. By increasing the temperature to -20 °C, a 60° angular deflection is obtained after 5 min. These results are in agreement with the DMA results and show that in the presence of EMTFSI, actuation is still possible down to at least -30 °C.

4. Conclusion

In this study conducting IPN based actuator architecture has been demonstrated and leads to a bending deformation with different salts and solvents as electrolytes. The device is made of poly(3,4-ethylenedioxythiophene) and a PEO/PB IPN. The dispersion of the conducting polymer in this device follows a symmetrical decrease from both outside faces towards the centre. This gradient distribution depends on the mean PEDOT content into the actuator, consequently the PEDOT mass quantity loaded in the conducting IPN was adjusted in order to get the electrical resistance along the surface at least 1000 times higher than the in the bulk. In this case, the structure is similar to that of a typical three-layer actuator.

The presence of RTIL (EMITFSI) in the conducting IPN decreases the alpha relaxation temperature from -45 to -70 °C showing that EMITFSI does not crystallize and still has a plasticizing effect. Consequently, the actuator can be operated in open air at low temperature (-30 °C), since EMITFSI can be used as electrolyte at this temperature.

References

- Groenendaal L, Jonas F, Freitag D, Pielartzik H, Reynolds JR. Adv Mater 2000;12:481–94.
- [2] Nalwa HS, editor. Handbook of organic conductive molecules and polymers. Conductive polymers: synthesis and electrochemical properties, vol. 2. New York: Wiley; 1997.
- [3] De Paoli M-A. Conductive polymer blends and composites. In: Nalwa HS, editor. Handbook of organic conductive molecules and polymers. Conductive polymers: synthesis and electrochemical properties, vol. 2. New York: Wiley; 1997. p. 773–95.

- [4] Sperling LH, Klempner D, Utracki LA, editors. Interpenetrating polymer networks. Washington, DC: American Chemical Society; 1991.
- [5] Dassonville P, Vidal F, Randriamahazaka H, Chevrot C, Teyssié D. Synth Met 2002;128:197.
- [6] Jeevananda T, Begum M, Siddaramaiah. Eur Polym J 2001;37:1213.
- [7] Gangopadhyay R, De A. Metals 2002;132:21.
- [8] Baughman RH. Synth Met 1996;78:339-53.
- [9] Smela E. Adv Mater 2003;15(6):481.
- [10] Vandesteeg N, Madden PGA, Madden JD, Anquetil PA, Hunter IW. Proc SPIE Conf Smart Struct Mater 2003;5051:349.
- [11] Chen X, Xing K-Z, Inganäs O. Chem Mater 1996;8:2439.
- [12] Vidal F, Popp J-F, Plesse C, Chevrot C, Teyssié D. J Appl Polym Sci 2003;90:3569.
- [13] Vidal F, Popp J-F, Chevrot C, Teyssié D. Proc SPIE Conf Smart Struct Mater 2002;4695:95.
- [14] Madden JD, Cush RA, Kanigan TS, Brenan CJ, Hunter IW. Synth Met 1999;105:61.
- [15] Sansiñena J-M, Olazàbal V. In: Bar-Cohen Y, editor. Electroactive

polymer actuator as artificial muscles. Bellingham: SPIE; 2001. p. 193-221 [Chapter 7].

- [16] Vidal F, Plesse C, Teyssié D, Chevrot C. Synth Met 2004;142:287.
- [17] Randriamahazaka H, Plesse C, Vidal F, Gauthier C, Chevrot C, Teyssié D. Proc SPIE Conf Smart Struct Mater 2004;5385:294.
- [18] Bonhôte P, Dias A-P, Papageorgious N, Kalyanasundaram K, Grätzel M. Inorg Chem 1996;35:1168.
- [19] Uchida M, Xu C, Le Guilly M, Taya M. Proc SPIE Conf Smart Struct Mater 2002;4695:57.
- [20] Vidal F, Popp JF, Chevrot C, Teyssié D. Proc SPIE Conf Smart Struct Mater 2002;4695:95.
- [21] Madden JD, Cush RA, Kanigan TS, Hunter IW. Synth Met 2000;113: 185.
- [22] Lu W, Fadeev AG, Qi B, Smela E, Mattes BR, Ding J, et al. Science 2002;297:983.
- [23] Zhou D, Spinks GM, Wallace GG, Tiyapibonchaiya C, MacFarlane DR, Forsyth M, et al. Electrochim Acta 2003;48:2355.
- [24] Ding J, Zhou D, Spinks G, Wallace G, Forsyth S, Forsyth M, et al. Chem Mater 2003;15:2392.