ELECTRONIC SUPPORTING INFORMATION

# Conducting polymer artificial muscle fibres: toward an open-air linear actuation

Cedric Plesse, Frederic Vidal, Dominique Teyssié and Claude Chevrot

*Laboratoire de Physico-chimie des Polymères et des Interfaces, Université de Cergy-Pontoise, Neuville sur Oise, 95031 Cergy-Pontoise, France*

## Experimental Section

*Chemicals.* Methoxy poly(ethylene glycol) methacrylate (PEGM, $M_w$ = 300 g.mol$^{-1}$), poly(ethylene glycol) dimethacrylate (PEGDM, $M_w$ = 875 g.mol$^{-1}$), α, ω dihydroxy poly(ethyleneoxide) (PEOdiOH, $M_w$ = 1500 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 be used. Toluene (Carlo-Erba), methanol (Carlo-Erba), anhydrous iron III chloride (Acros), and Desmodur® N3300 (5.2 10$^{-3}$ mole of NCO for 1g of Desmodur®) (Bayer) were used without further purification. Azobisisobutyronitrile (AIBN) (Aldrich) was recristallized in methanol.

*Synthetic procedure of* ***Poly(ethyleneoxide) / Poly(ethyleneoxide) IPN SPEs (PEO/PEO IPN).*** PEO/PEO IPN is prepared according to the following procedure. Typically, AIBN is used for methacrylate function initiation of PEGDM and PEGM in the formation of the first PEO network. Desmodur® N3300 is the PEOdiOH crosslinker and DBTDL is the catalyst of the reaction between NCO and OH functions. The mixture of PEGDM, PEGM and PEOdiOH (0.1 / 0.4 / 0.5 weight proportions) were poured into a flask. Desmodur® (1.1 eq / OH functions) and AIBN (3 wt% of the PEGDM-PEGM mass) are then added to the mixture. 3,4-ethylenedioxythiophene (EDOT) (120wt% versus total PEO weight) is then added to ensure the homogeneity of the mixture. The solution was stirred under argon atmosphere during 30 minutes. DBTDL was added (10 mol% / OH functions) and the mixture was poured into a mould. The mould was heated at 60 °C for 3 hours and post-cured for 1 hour at 80 °C. The EDOT swollen hollow IPN fibers are stored at 5°C under argon.

*Synthetic procedure of* ***Conducting Polymer Based IPN Actuator.*** The EDOT polymerization is performed by chemical oxidation with iron chloride aqueous solution under microwave

irradiation, according to the procedure described by J. Citerin et al. (PhD, unpublished results). The standard procedure is the following: EDOT swollen hollow IPN fibers were immersed into test tubes containing 5 mL of a $FeCl_3$ aqueous solution (1.5 $mol.L^{-1}$).

The test tubes are introduced in a beaker containing 1.3L water (figure 1) and placed into a microwave oven (850 W).

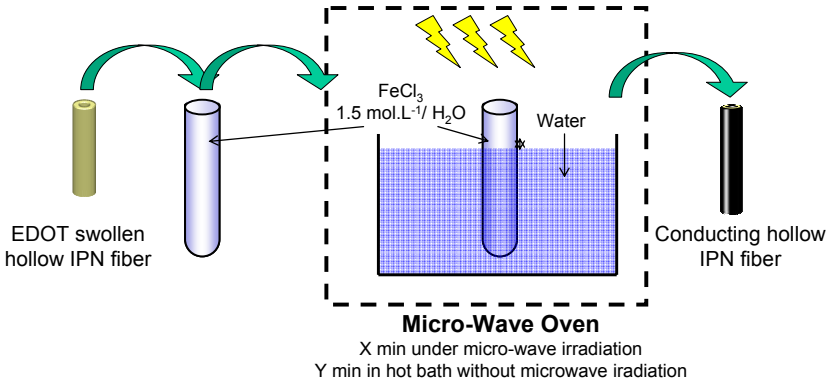


Figure 1: schematic representation of EDOT polymerization procedure under microwave irradiation.

The increase of the temperature, under microwave irradiation, leads to an increase of the polymerization rate.

The sample is heated during 7 to 9 minutes and left 2 additional minutes in the hot bath but without microwave power. The conducting hollow IPN fiber in the iron chloride solution was immersed in cold water in order to stop the polymerization process.

The conducting IPN fiber is washed several times with methanol until the solvent remains colorless i.e. the $FeCl_3$ excess is removed. The actuator surface is then wiped off with filter paper and the two extremities are cut off. The conducting material is dried at 50°C under vacuum for 24 hours.

The conducting fibres (figure 2) are then soaked and stored in the chosen electrolyte, $LiClO_4$ 1 M propylene carbonate.

Actuator characterization

The electrical resistances values can give interesting informations about the PEDOT anisotropic dispersion inside the IPN matrix. The two terminal-technique with gold pressure contact was used for the room temperature resistance measurements. The measurement of the

resistance was performed along the outer surface ($R_{out}$), the inner surface ($R_{in}$) and across the thickness or bulk ($R_b$) with a KEITHLEY 197 Autoranging microvolt DMM (Figure 2).

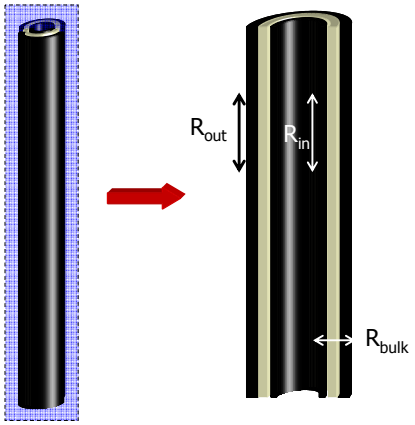


Figure 2: schematic representation of the vertical cross-section of the actuator.

Whatever the polymerization time, the outer and inner resistances values, $R_{out}$ and $R_{in}$, vary from 10 to 60 Ω. However, the bulk resistance $R_b$ is at least 100 times higher than the resistance measured along the surfaces and decreases when the polymerization time increases (Figure 3). This result indicates that the EDOT polymerization occurs initially from the (inner and outer) surfaces of the fibre and then, proceeds progressively through the depth of the IPN. For longer polymerisation time, percolation occurs with bulk resistances lower than 20 Ω.

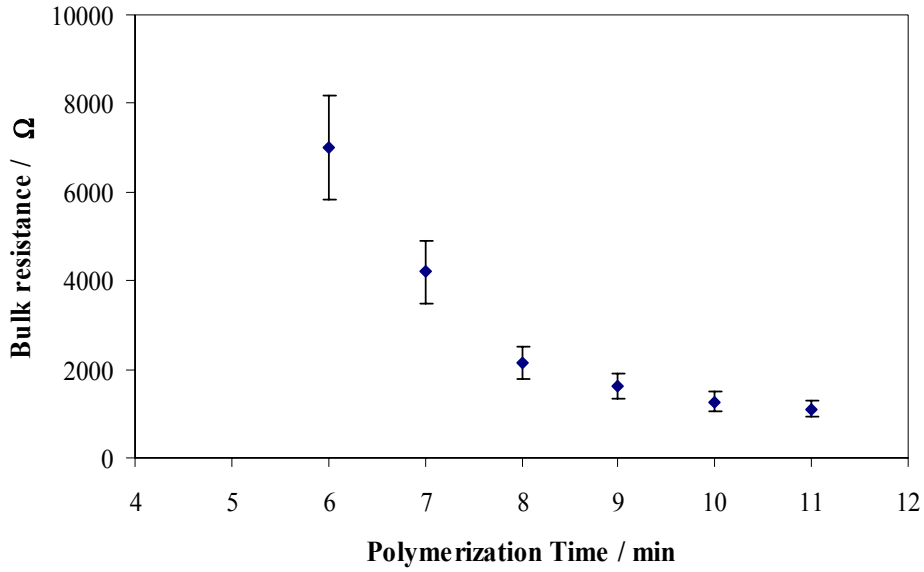


Figure 3: evolution of the bulk resistance, i.e. the resistance measured between the outer and the inner PEDOT electrodes, as a function of the polymerization time (X) under microwave irradiation followed by 2 additional minutes in hot bath without irradiation.

**Actuation Testing**

After the extremities of the fibres were cut off, the conducting IPNs were immersed in $LiClO_4$ 0.1 $mol.L^{-1}$ / propylene carbonate for one hour at room temperature. The conducting IPNs were then maintained vertical with a steel electrode inserted in the inner hole of the fiber, and another one surrounding the outer part, in order to ensure electrical contact with the potentiostat. The linear actuation response was obtained by applying a 3V square potential wave.

The relative deformation of the fiber was recorded under isotonic mode (0.1 N) with a laser displacement sensor (ILD 1401-5, Micro-Epsilon) fitted to a computer. The force output was measured with an analytical balance (Kern) and recorded through Regressi® software in isotonic mode under different preload forces (0.1 to 1.7 N).

As seen in figure 6 of the article, the output force still increases as a function of the time, even after 100 seconds. This phenomenon can be related to a creeping process. Indeed, this curve corresponds to a linear actuator with a high preload force (1.7 N).. As it can be seen below on figure 4, under lower preload forces (0.44 N), the creeping behaviour of the hollow IPN fibre actuator almost disappears.

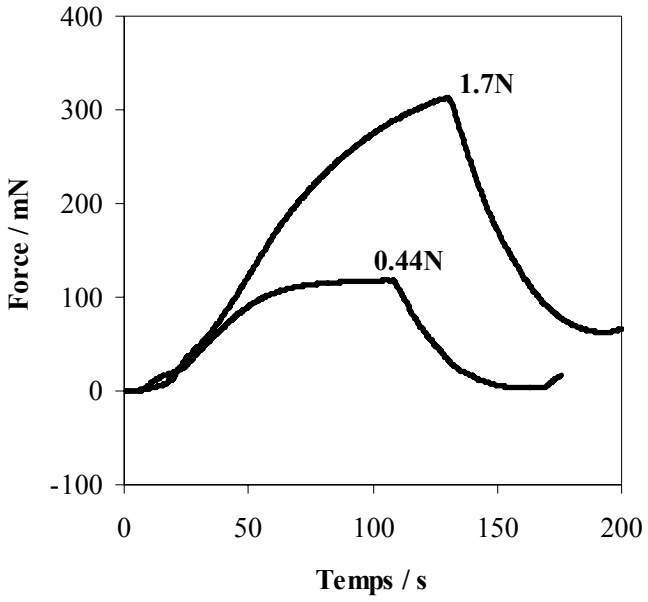


Figure 4: output force of a PEO/PEO/PEDOT IPN fibre actuator. ΔE = +/- 3V. Preload force: (A) 1.7 N, (B) 0.44 N.