

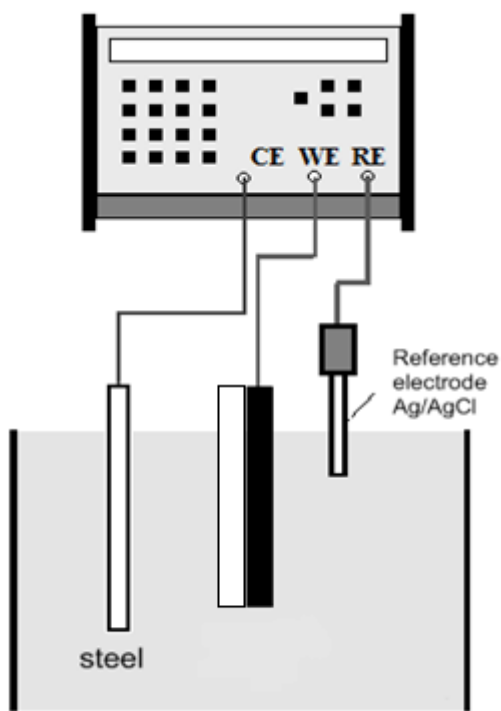
## Electronic Supplementary Information

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

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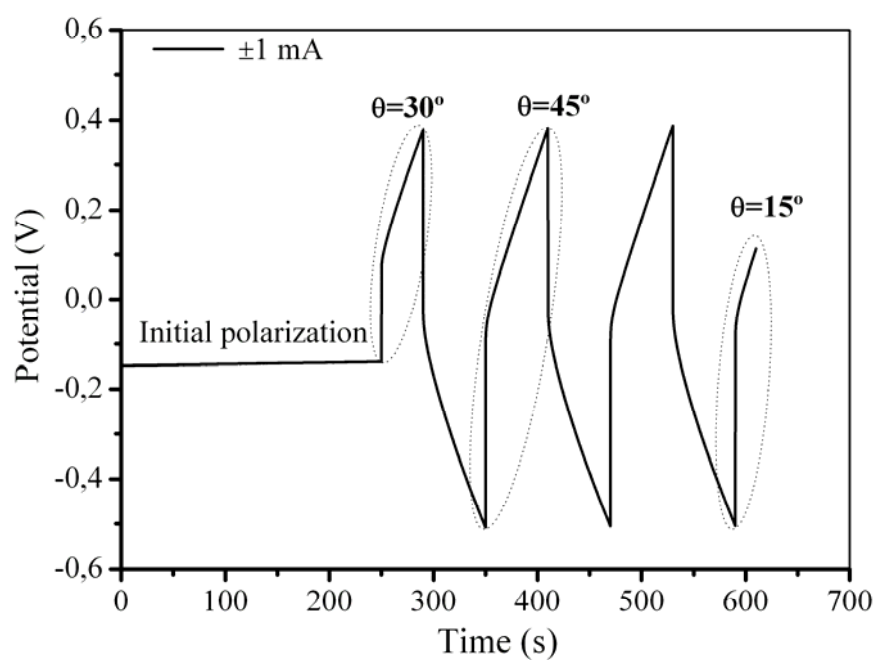
### Supplementary Information S1

Scheme of the experimental configuration (WE, working electrode; CE, counter electrode; RE, reference electrode) used to study the electrochemical actuation and sensing behaviour of the by-layer actuators



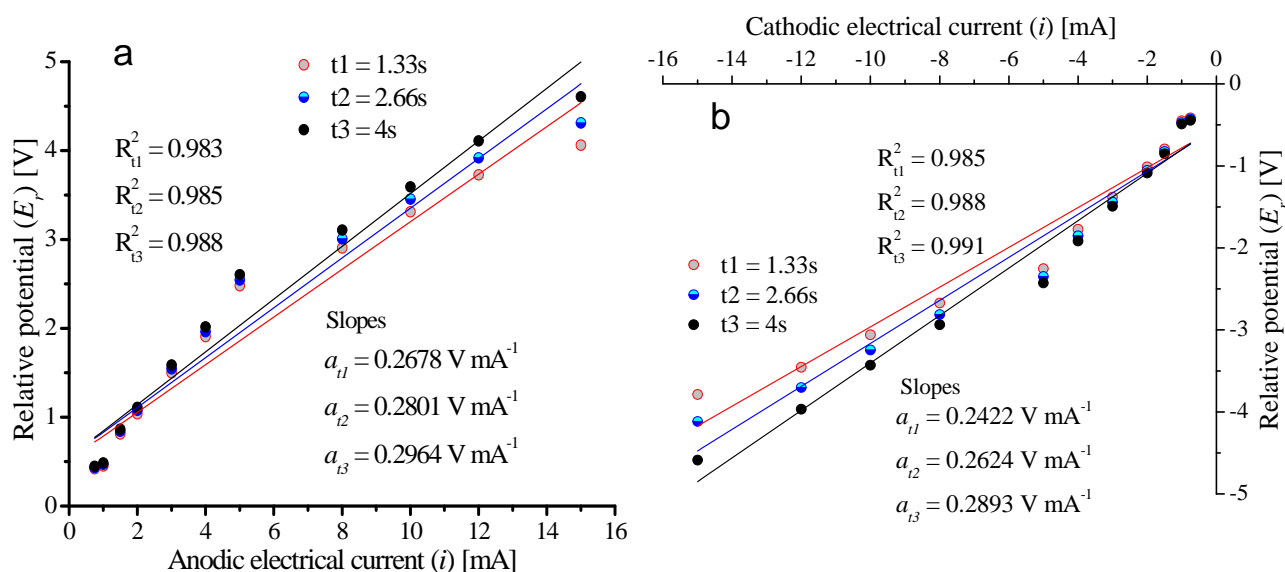
## Supplementary Information S2

Chronopotentiogram obtained by applying an initial polarization of  $-0.01$  mA and consecutive square current waves of  $\pm 1$  mA in aqueous solution of  $0.1\text{M LiClO}_4$ .



## Supplementary Information S3.a & S3.b

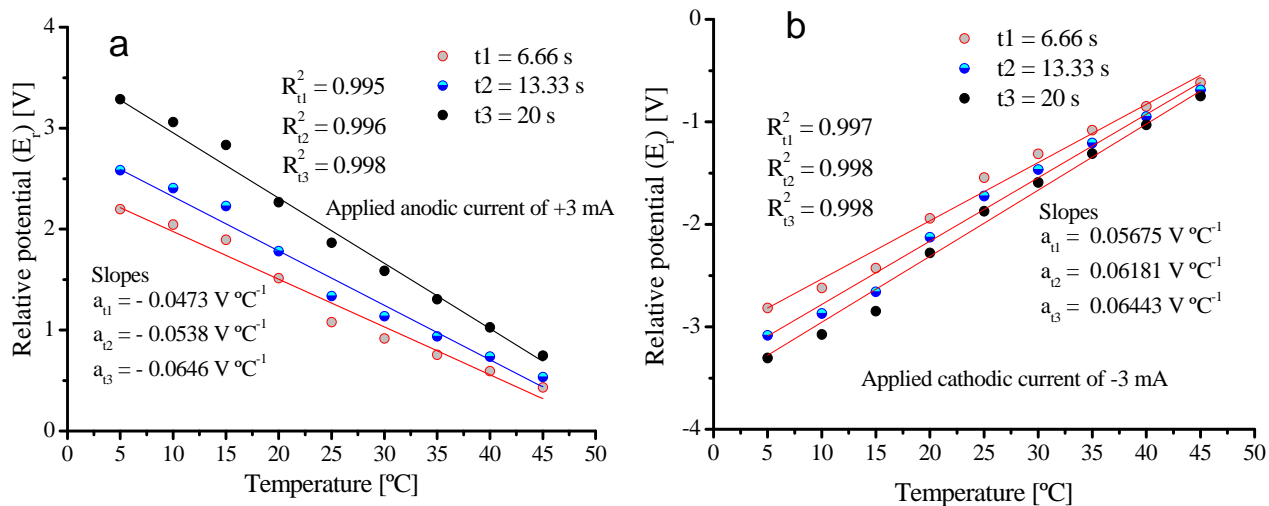
The potential evolution at different currents during the chronopotentiometric studies indicated in Fig 5a and Fig.5b at different time periods corresponding to  $t_1 = 1.33$  s,  $t_2 = 2.66$  s and  $t_3 = 4$  s which respectively produced angular movements of  $15^\circ$ ,  $30^\circ$  and  $45^\circ$ . S3a and S3b represents the responses for anodic and cathodic processes respectively.



From the chronopotentiometric responses obtained by flow of  $\pm 15$  mA of current through the three layers device, the muscle potential after describing three different angular movements of  $\pm 15^\circ$ ,  $\pm 30^\circ$ , and  $\pm 45^\circ$ , respectively, were determined. Those angular movements correspond to three different periods of current flow,  $t_1 = 1.33$  s,  $t_2 = 2.66$  s and  $t_3 = 4$  s, respectively. Fig.S3a and S3b respectively show the linear relationship of the muscle potential after describing  $\pm 15^\circ$ ,  $\pm 30^\circ$ , and  $\pm 45^\circ$  at different applied anodic or cathodic currents, respectively. Slopes represent that the muscle potential increases when the magnitude of the driving current increases by one unit. Slopes are found to increase with the period of time of the applied current.

## Supplementary Information S4a and S4b

The muscle potential after different periods of current flow:  $t_1 = 6.66$  s,  $t_2 = 13.33$  s and  $t_3 = 20$  s, which respectively produced angular movements of  $15^\circ$ ,  $30^\circ$  and  $45^\circ$  at different temperatures and obtained from the chronopotentiometric results in fig 6a and fig.6b.



## Supplementary Information S5a and S5b

The potential evolution at different electrolyte concentrations during the chronopotentiometric studies as indicated in fig 7a and fig.7b at different times corresponding  $t_1 = 6.66$  s,  $t_2 = 13.33$  s and  $t_3 = 20$  s. which respectively produced angular movements of  $15^\circ$ ,  $30^\circ$  and  $45^\circ$

