Electronic Supplementary Information

Electrochemical characterization of the stimuli-response of surface-immobilized elastin-

like polymers

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S1: SDS-PAGE



Figure S1. Coomassie-stained SDS-PAGE analysis of purified I40 polymer consistent with the predicted molecular weight of 17.9 kDa.

S2: I40 Polymer – Cysteine Blocking Validation:

Verification of the thiol alkylation of I40-Blocked was achieved using Ellman's Reagent 5,5'dithiobis-(2-nitrobenzoic acid), often referred to as DTNB, to quantify the number of available thiols. The conjugate base of the free sulfhydryl groups (R-S') cleaves the disulfide bond on the DTNB to form a mixed disulfide and 2-nitro-5-thiobenzoate (TNB) a reaction product yellow in color which can be measured using spectroscopy.

 $300 \ \mu\text{L}$ of a prechilled solution of 1.0 mg/mL I40 or I40-Blocked in 10% DMSO was mixed with 100 μL of 4 mg/mL of DTNB. The reaction proceeded on ice for 15 minutes before measuring using UV-Vis. As seen in **Figure S3**, there is a decrease in absorbance for I40-Blocked polymer compared to I40 indicating a decrease in the number of available thiols on the polymer after alkylation with iodoacetamide.



Figure S2. Comparison of I40 to I40-Blocked after reaction with DTNB.

S3: Residual curves confirming appropriate interpretation of data

Residual fractional error curves evaluating the quality of fit of the Randle's circuit model to the impedance data. All fits (I40, I40-Blocked and Unmodified) show even distribution over the frequency range with a lack of major trends, which indicates the Randle's circuit model is appropriate to fit the data (**Figures S3** and **S4**).



Figure S3. Residual fractional error curve evaluating quality of fit for an I40 modified electrode.



Figure S4. Residual fractional error curve evaluating quality of fit for an unmodified electrode and an I40-Blocked modified electrode.

S.4: Attachment experimental error

Six different electrodes were modified with I40 (**Figure 5**). The charge transfer is calculated for each attachment (N=6) with fit error at $\pm 0.1 \text{ k}\Omega$ (**Table S1**).

Table S1. Charge transfer resistance values for each electrode in **Figure 5** showing surface modification reproducibility with an average charge transfer resistance value of 12.7 ± 1.4 k Ω s

	$\mathbf{R}_{\mathbf{ct}} \left(\mathbf{k} \mathbf{\Omega} \right) \pm 0.1$
Electrode 1	17.1
Electrode 2	13.6
Electrode 3	15.0
Electrode 4	12.0
Electrode 5	12.8
Electrode 6	11.0

S5. I40 Modified Gold - 10 mM [Fe(CN)6]^{3-/4-} Impedance Response

The impedance response from an I40 modified gold electrode was obtained after soaking overnight in 0.0 M NaCl. The collapsed state of the ELP was triggered by soaking the I40 modified Au electrode in 3.0 M NaCl for two hours; an increase in charge transfer resistance is observed. The I40 modified electrode was then left to soak in the 10 mM $[Fe(CN)_6]^{3-/4}$ redox couple solution overnight before measuring the impedance response. The impedance response after exposing the I40 modified surface to (1) 10 mM $[Fe(CN)_6]^{3-/4-}$ and (2) 0.0 M NaCl are similar indicating the ELP have a similar surface response to these two solutions (**Figure S5**). This data verifies that the molarity of the redox couple solution is sufficiently low to not influence the stimuli-response of surface-immobilized I40.



Figure S5. Impedance response obtained after soaking an I40 modified electrode in the redox couple solution

S6. I40 Modified Gold – Stimulus Response

Stimuli-response from two unique I40 modified electrodes exposing to 0.0 M NaCl, 0.5 M NaCl, 0.75 M NaCl, and 1.0 M NaCl (**Figure S6**). The data shows the run-to-run reproducibility of the stimuli-response across different electrodes, an intermediate state at 0.5 M NaCl of both electrodes, and a saturation in response over 0.75 M NaCl.



Figure S6. Replicate data of the impedance response from an I40 modified electrode after exposure to 0.0 M NaCl-1.0 M NaCl. An intermediate state between the maxima and minima is observed at both electrodes under a 0.5 M NaCl condition.