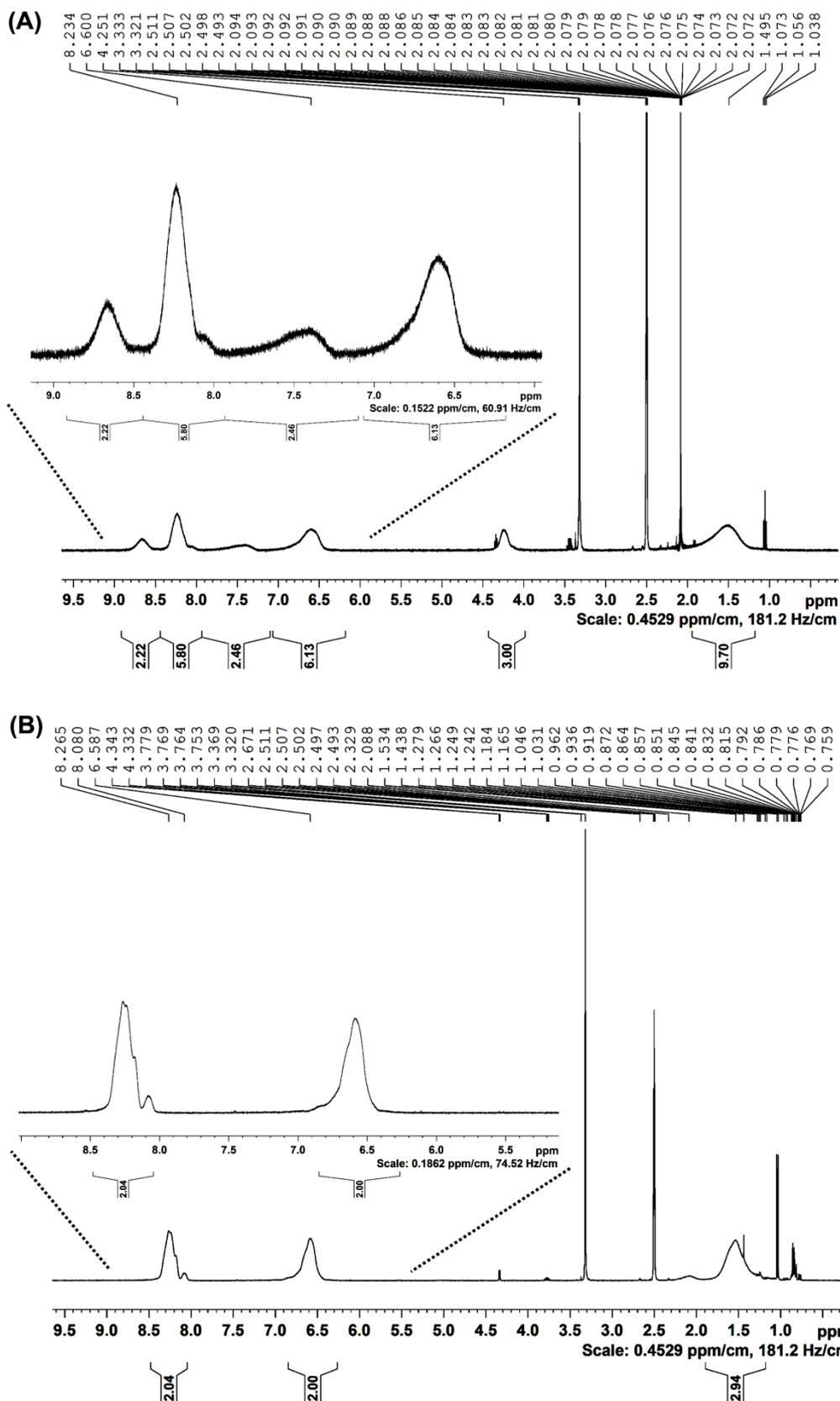


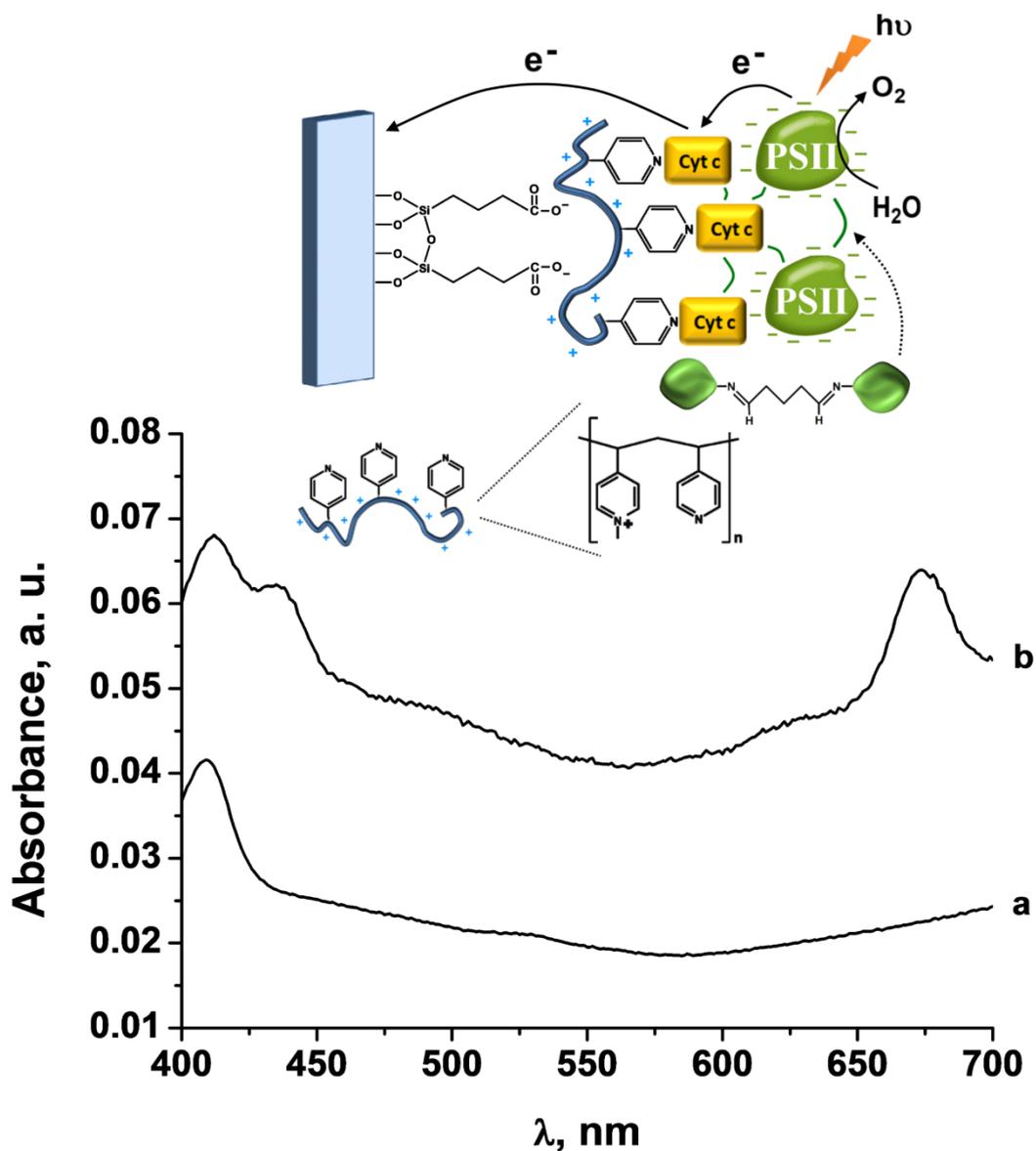
Supplementary Information for the paper:

**Cytochrome c-Coupled Photosystem I and Photosystem II (PSI/PSII)  
Photo-bioelectrochemical Cells**

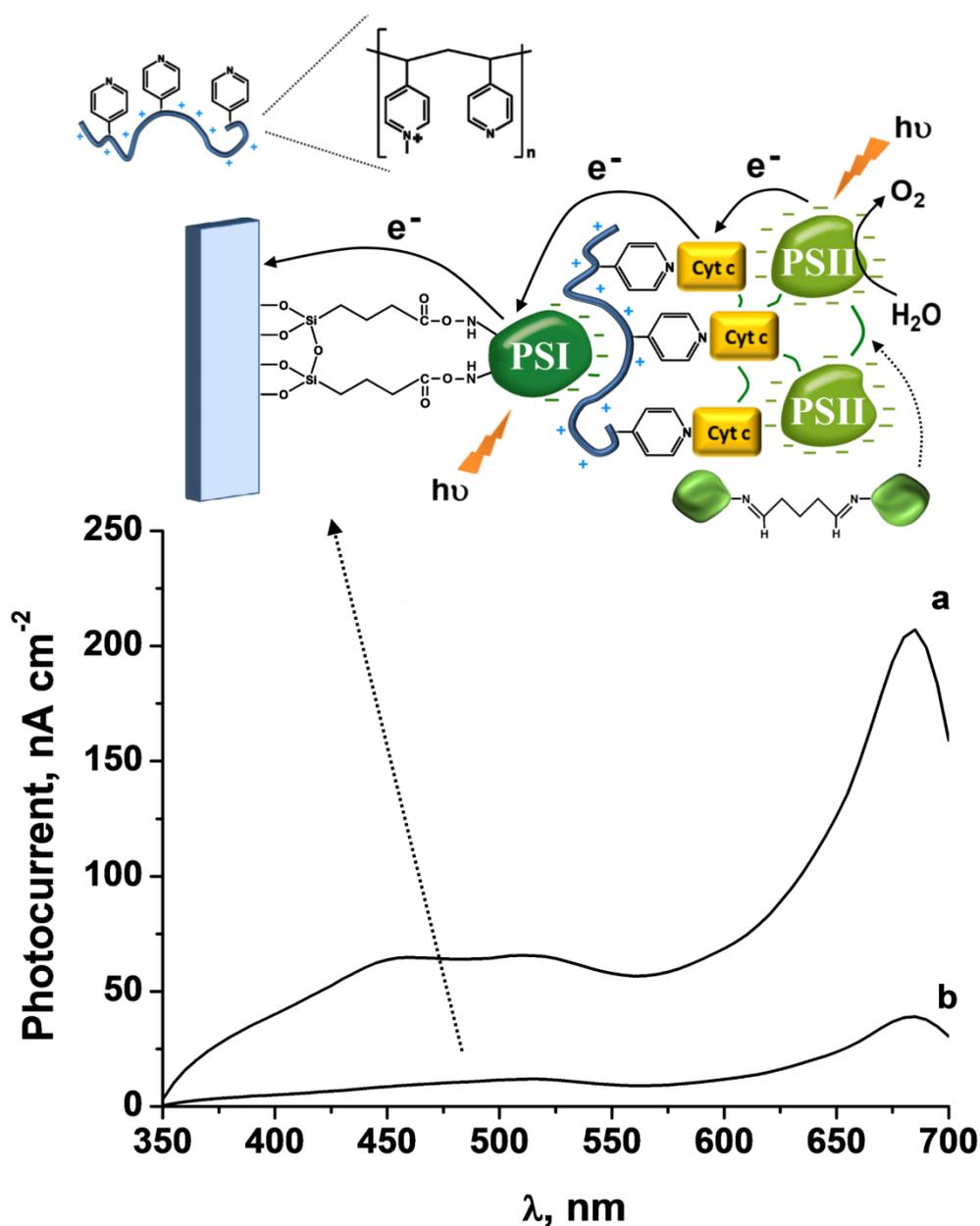
*Ariel Efrati, Ran Tel-Vered, Dorit Michaeli, Rachel Nechushtai and Itamar Willner*



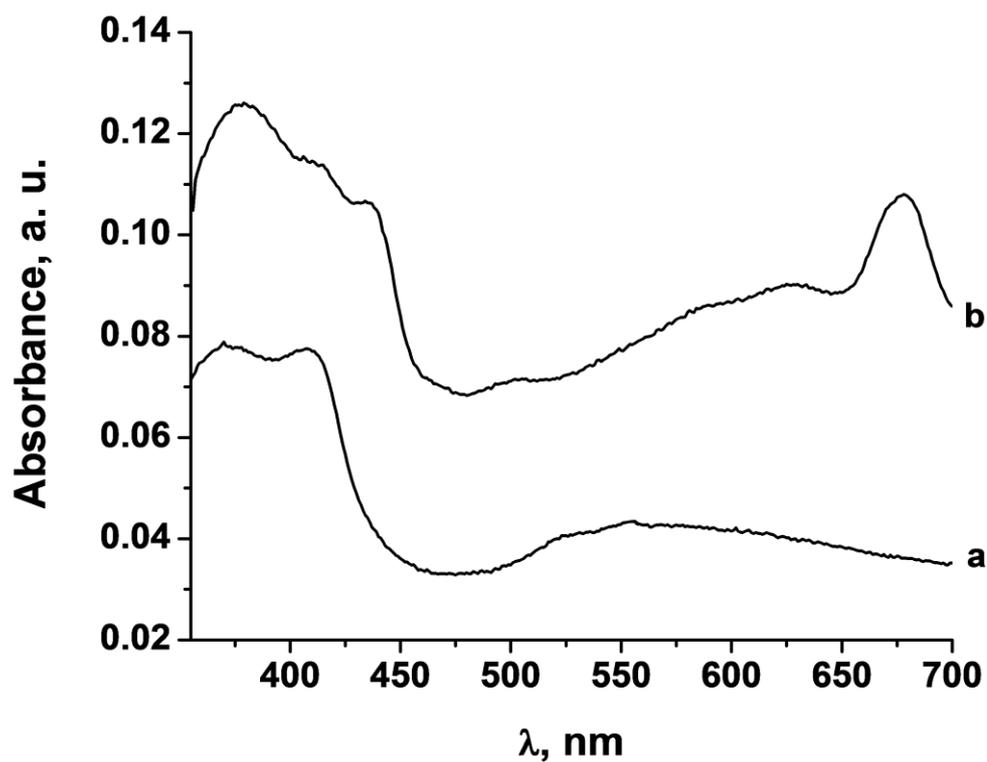
**Figure S1.** NMR spectra obtained for: (A) The polyvinyl pyridine/methyl pyridinium copolymer, and (B) The polyvinyl pyridine polymer.



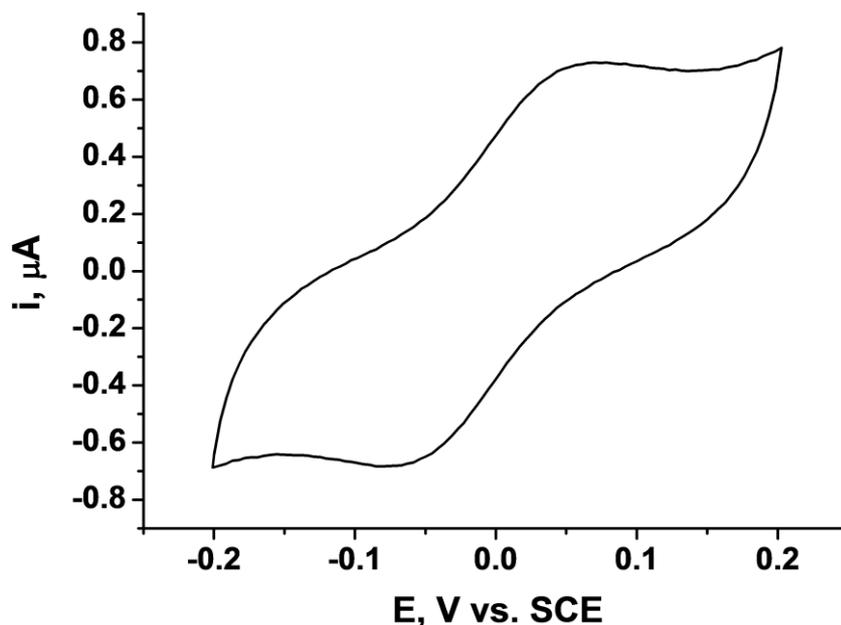
**Figure S2.** Absorption spectra corresponding to the stepwise assembly of the Cyt *c*/PSII photoactive composite on a carboxypropyl siloxane-modified ITO electrode: (a) following the interaction of the surface with the polyvinyl pyridine/methyl pyridinium copolymer-Cyt *c* layer; (b) upon the interaction of the resulting assembly with PSII, and crosslinking with glutaric dialdehyde.



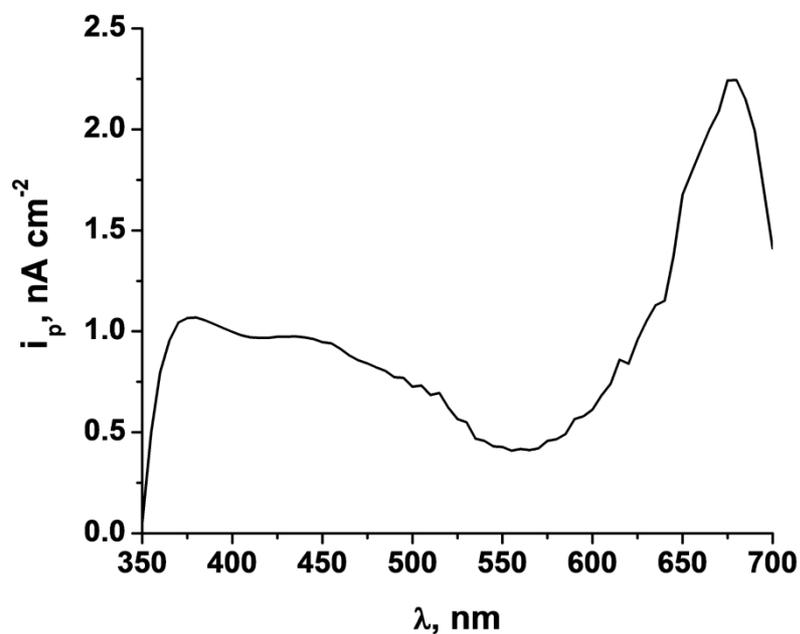
**Figure S3.** Photocurrent action spectra recorded at open circuit potential: (a) The aligned Pt NC-PSI/Cyt c/PSII composite-modified ITO electrode (a monolayer of (3-mercaptopropyl) trimethoxysilane is used as a linker between the Pt NC and the ITO). (b) The non-aligned PSI/Cyt c/PSII composite-modified ITO electrode (a monolayer of carboxypropyl siloxane is used to couple the lysine residues of the PSI to the ITO surface). The measurement was performed in a phosphate buffer (pH=7.2, 0.1 M).



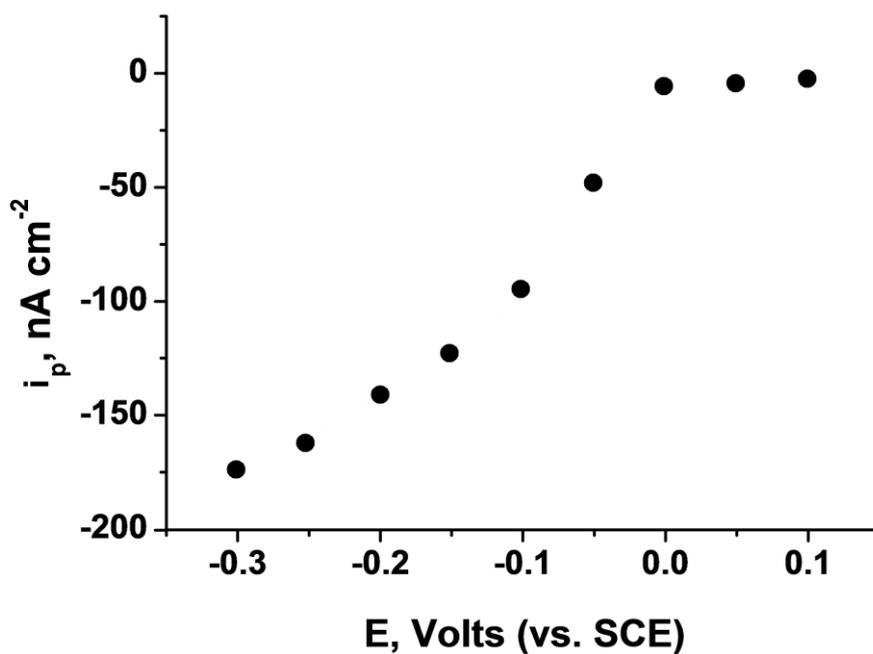
**Figure S4.** Absorption spectra corresponding to the stepwise assembly of the Cyt c./PSI photoactive composite on a carboxypropyl siloxane-modified ITO electrode: (a) following the interaction of the surface with the polyvinyl pyridine/methyl pyridinium copolymer-Cyt. c layer; (b) upon the interaction of the resulting assembly with PSI, and crosslinking with glutaric dialdehyde.



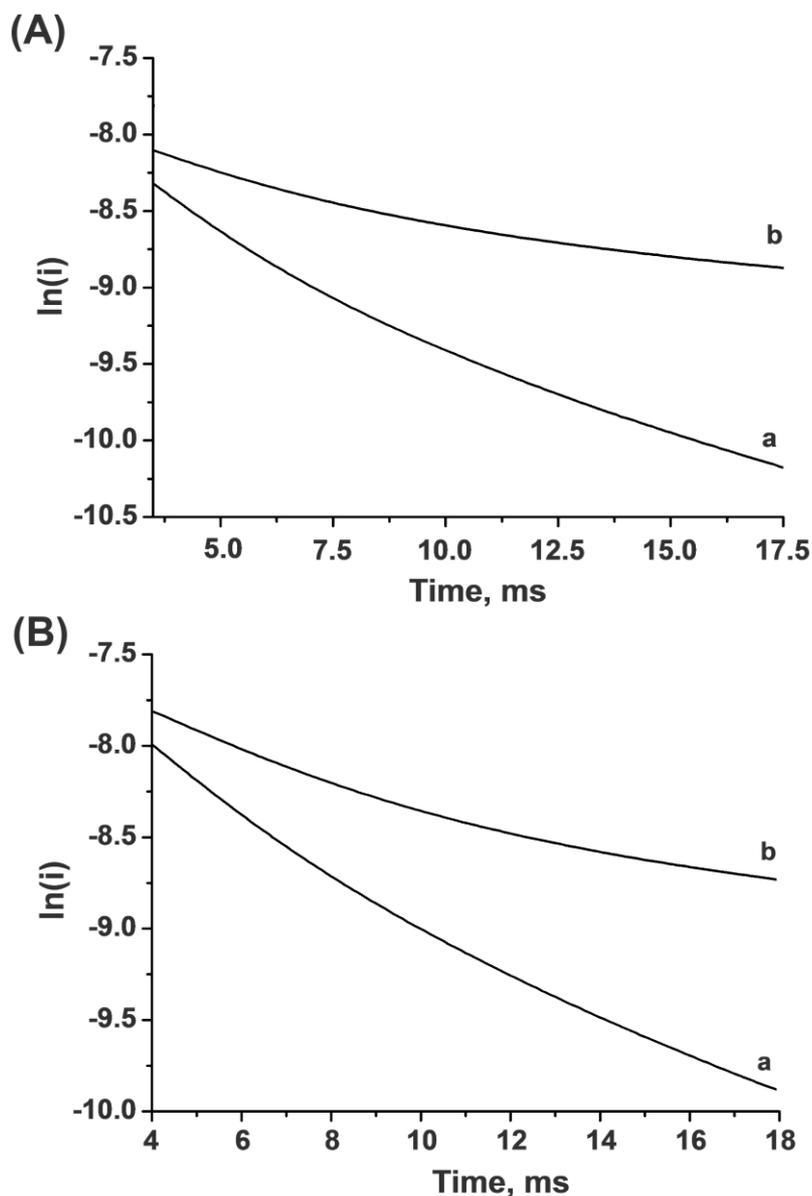
**Figure S5.** Cyclic voltammogram corresponding to carboxypropyl siloxane-modified ITO electrode, following its interaction with the polyvinyl pyridine/methyl pyridinium copolymer-Cyt. c complex. Measurement was performed in phosphate buffer (pH=7.2, 0.1 M) at a scan rate of  $50 \text{ mV s}^{-1}$ .



**Figure S6.** Photocurrent action spectrum recorded for a Cyt c./PSI composite-modified ITO electrode at open circuit potential. The measurement was performed in phosphate buffer (pH=7.2, 0.1 M) that contained  $3 \times 10^{-4} \text{ M MV}^{2+}$ .



**Figure S7.** Dependence of photocurrent intensity, at  $\lambda=680$  nm, on the bias potential applied on the Cyt c./PSI composite-modified electrode. Measurement was performed in phosphate buffer (pH=7.2, 0.1 M).



**Figure S8.** (A) Chronoamperometric responses corresponding to Cyt. c monolayer-modified electrodes. The Cyt. c was adsorbed on a Au surface functionalized with a monolayer of: (a) 4-Mercaptopyridine, and (b) Mercaptopropionic acid. (B) Chronoamperometric responses corresponding to Cyt. c adsorbed on: (a) Polyvinyl pyridine/methyl pyridinium-, and (b) Polylysine-modified Au surfaces. The polymers were first deposited on mercaptopropionic acid-functionalized Au slides, and following the interaction of the polymer-modified electrodes with Cyt. c, crosslinking with glutaric dialdehyde was employed. For all measurements a potential step between -0.2 V and 0.2 V vs. SCE was applied. Measurements were performed in phosphate buffer (pH=7.2, 0.1 M).