

**Electronic Supporting Information for the article:**

**A Coupled Electrochemical/Photochemical Switchable Information Processing System Using a Monolayer-Functionalized Electrode**

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**Preparation of compound (1):**

1,2-bis[2-methyl-5-(4-pyridyl)-3-thienyl] cyclopentene was prepared according to ref. 12. The prepared compound, 15 mg, was dissolved in 1 mL of solvent (70% acetone and 30% DMF) and 200 mg of bromoacetic acid was added to the solution. After stirring for 5 min, the mixture was heated at 60°C for 24 h in the dark. The solution was then allowed to cool at room temperature and cold acetone was added dropwise until the solid precipitated completely. The precipitate was then collected by centrifugation (2800 rpm, 4°C, 15 min) and washed with cold acetone twice.

**Electrode modification:**

The (2a)-modified electrode was modified as depicted in Scheme 1. A cysteamine monolayer was assembled on the Au plates by immersing the electrode overnight in a 10<sup>-2</sup> M cysteamine aqueous solution. The electrode was then put in a HEPES buffer solution, 30 mM, pH=7.3, containing (1a), 2 × 10<sup>-3</sup> M and EDC, 5 × 10<sup>-2</sup> M for 3 hours.

**Electrochemistry:**

Electrochemical experiments were performed using a saturated calomel reference electrode (SCE) separated from the rest of the solution by a Luggin capillary. The auxiliary electrode was a glassy carbon cylinder. The working electrodes were Au-coated (250 nm) glass slides (Evaporated Coatings Inc. USA). The surface area of the working electrodes in contact with the solution was 0.7 cm<sup>2</sup>, and their roughness factor was estimated to be ca. 1.2. The support electrolyte was phosphate buffer, 0.1 M, pH=7.2. All experiments were performed in the dark at ambient temperature under an argon atmosphere. The electrochemical investigations were performed using an Eco Chemie Autolab potentiostat-galvanostat PGSTAT12 connected to a computer.

The bulk electrolysis was conducted, in phosphate buffer solution, pH=7.2, under stirring and using an unmodified Au-coated glass slide as the working electrode.

**Light irradiation:**

The irradiation source was a 300 W Xe lamp (Oriel, model 6258) equipped with a monochromator (Oriel, model 74000). The power of the lamp at 570 nm was measured to be 2.98 mW.