Supplementary information

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Sample preparation. Si wafers with 300 nm SiO\(_2\) were purchased from University Wafer, Inc., USA. Five nm of titanium (adhesive layer) and 150 nm of nickel were evaporated on the silicon wafers. After evaporation, the samples were cut and kept in a nitrogen environment until use. A small beaker with a defined area of 0.78 cm\(^2\) was sealed on the nickel surface to ensure a well-defined area. The beaker was sealed from the outside using glue. In this way, there is no contact between the glue and the liquid (as shown in the Figure S1).

**Figure S1:** A picture of a sealed beaker glued on the Ni surface. The glue is used only from the outside; therefore, there is no contact between the glue and the active Ni surface.

Both wild-type and D96N bR were prepared in carbonate buffer (5 µM in 50 mM sodium carbonate buffer). Different amounts of wild-type and D96N bR were physisorbed on the
surface using the drop-casting method and left to dry in a vacuum of $10^{-2}$ Torr for 48 hours.

Three types of samples were prepared, each three times. The types are as follows: a. Different amounts of drop-casted bR (WT and D96N) on Ni with 0.78 cm$^2$ active area. b. bR deposition using an electric field. c. Chronoamperommetry was measured both on the same samples as used for the cyclic voltammetry and on new fresh samples in order to reduce the measurement time and to avoid Ni oxidation. In this paper the chronoamperometry results represent the freshly made samples.

**Electrochemical measurements.**

Cyclic voltammetry and chronoamperometry were measured using a potentiostat (SP-200, Bio-Logic LTD). The electrochemical measurements were performed under ambient conditions in the dark and during illumination at 532 nm. The laser used in this work is a Temperature Controlled Laser Diode Mount (TCLDM9, 3mW power, ThorLabs, USA). The diameter of the spot is 5.6 millimeters. When light shines on the electrochemical cell, the light is scattered and covers the whole area of the active surface. The working electrode is nickel; underneath, a magnet is placed whose direction can be flipped (0.35 T). The counter electrode is platinum and the reference electrode used is KCl-saturated calomel. The electrolyte used in all electrochemical measurements consists of a 10 mM tris(hydroxymethyl)aminomethane (TRIS) buffer with 50 mM NaCl at pH 9, with the addition of a redox couple; a basic pH was chosen in this work in order to increase the M's intermediate lifetime. The redox couple selected for the current study was $K_d[Fe(CN)_6]/K_3[Fe(CN)_6]$ (Fe$^{2+}$/Fe$^{3+}$) due to its robust chemical properties and because of our thorough knowledge of its thermodynamic, kinetic, and electrochemical parameters. The relevant chemical reaction is a reversible electrochemical equilibrium. The relevant standard potential is:

$E_0=+0.120$ V vs SCE (Saturated Calomel Electrode):

$$Fe(CN)_6^{3+}_{(solution)} + e^- = Fe(CN)_6^{2+}_{(solution)}$$
Cyclic voltammetry measurements were performed at the beginning to determine the oxidation and reduction potentials of each sample. The cyclic voltammetry measurements were performed with 5 mM Fe$^{2+}$/Fe$^{3+}$. The scan rate is 100 mV/s. The chronoamperometric measurements were done at the reduction potentials in three concentrations of the redox couple: 1 mM, 5 mM, and 10 mM. Different concentrations of redox couples were used in order to eliminate the effect of diffusion of the redox couple to the surface while applying oxidation or reduction potentials. Measuring with different redox concentrations indicated that the effect shown here is not related to the diffusion of the redox couple and can be seen even in samples with high concentration of redox in the electrolyte. These measurements aim to strengthen the conclusions of the measurements and to eliminate the effect of parallel processes.

All measurements were performed as a function of the direction of the magnetization of the Ni electrode in the dark and when illuminated at 532 nm. The magnetic field used in this work is 0.35 T.

To eliminate the possibility that the spin polarization effect comes from the Ni surface, cyclic voltammetry was applied to a clean Ni surface under the same conditions and with the same electrolytes as stated above. Figure S2 shows the cyclic voltammetry of a clean Ni surface in TRIS buffer and 5 mM Fe$^{2+}$/Fe$^{3+}$.

![Graph](https://via.placeholder.com/150)

**Figure S2**: CV curves of bare Ni in 5 mM K$_4$(Fe(CN)$_6$)/K$_3$(Fe(CN)$_6$) redox couple in TRIS buffer, pH 9, in a magnetic field (0.35 T) that is orthogonal to the surface of the working electrode. The scan rate is 100 mV/s.

Figure S3 shows cyclic voltammetry measurements of WT bR in the dark and during illumination at 532 nm. It can be clearly seen that the effect of light on WT bR cannot be
detected under these experimental conditions. The ratio in the current through the membranes containing the mutant and the WT bR is shown in Figure S4.

**Figure S3:** CV curves of WT bR in the dark and during illumination. The electrolyte used is 5 mM $K_4[Fe(CN)_6]/K_3[Fe(CN)_6]$ redox couple in TRIS buffer, pH 9, in a magnetic field (0.35T) that is perpendicular to the surface of the working electrode. The scan rate is 100 mV/s.

**Figure S4:** The current through the purple membrane containing the mutant D96N (black) and through the WT bR (red) measured for different concentrations of the deposition solution at the peak of the oxidation potential. The results are shown for a magnetic field pointing upwards. A similar trend was shown for the opposite magnetic field.
Membrane orientation

For estimating the samples' orientation, we deposited the bR using an applied electric field. Following the deposition step, we dried the samples and measured the CPD and the electrochemical signal. The electric field used in this work was ± 40 V/cm. All the samples were measured 3 times for statistical purposes.

Figures S5 and S6 present our results. The membrane was deposited in the presence of an electric field since it was previously shown (see for example: K. M. Bromley, A. J. Patil, A. M. Seddon, P. Booth, S. Mann, *Adv. Mater.* 2007, 19, 2433–2438) that the electric field induces the orientation of the bR sample. It was found that whereas in the case of the WT there is some effect of the field, its effect is smaller on the mutant orientation. However, when no field is applied, the CPD shows results similar to those obtained when a field is applied, so that the substrate is biased positively relative to the counter electrode. The reproducibility of CPD measurements for all samples indicates that the membrane is deposited when the more negative side is pointing towards the substrate.

![Figure S5: CPD measurements of WT and D96N deposited using an electric field of ± 40 V/cm. The results are normalized with the work function of the bare Ni. The amount of bR on the surface is equal in all the samples and is 3.53 (1000*µg/µL²).](image)
**Figure S6:** CPD measurements of WT and D96N bR deposited on Ni surfaces using the drop-casting method (without an electric field). The amount of bR on the surface varied.

**AFM measurements**

AFM measurements were performed on thin and thick drop-casted WT and D96N. Figure S7 shows that good surface coverage was obtained for all samples.

**Figure S7:** AFM measurements performed on Ni substrate coated with the purple membrane containing either the wild-type or mutant bR.