Supporting Information

Chromogenic Homo-Dinuclear Ruthenium(II) Monolayer as a Tunable Molecular Memory Module for Multibit Information Storage†

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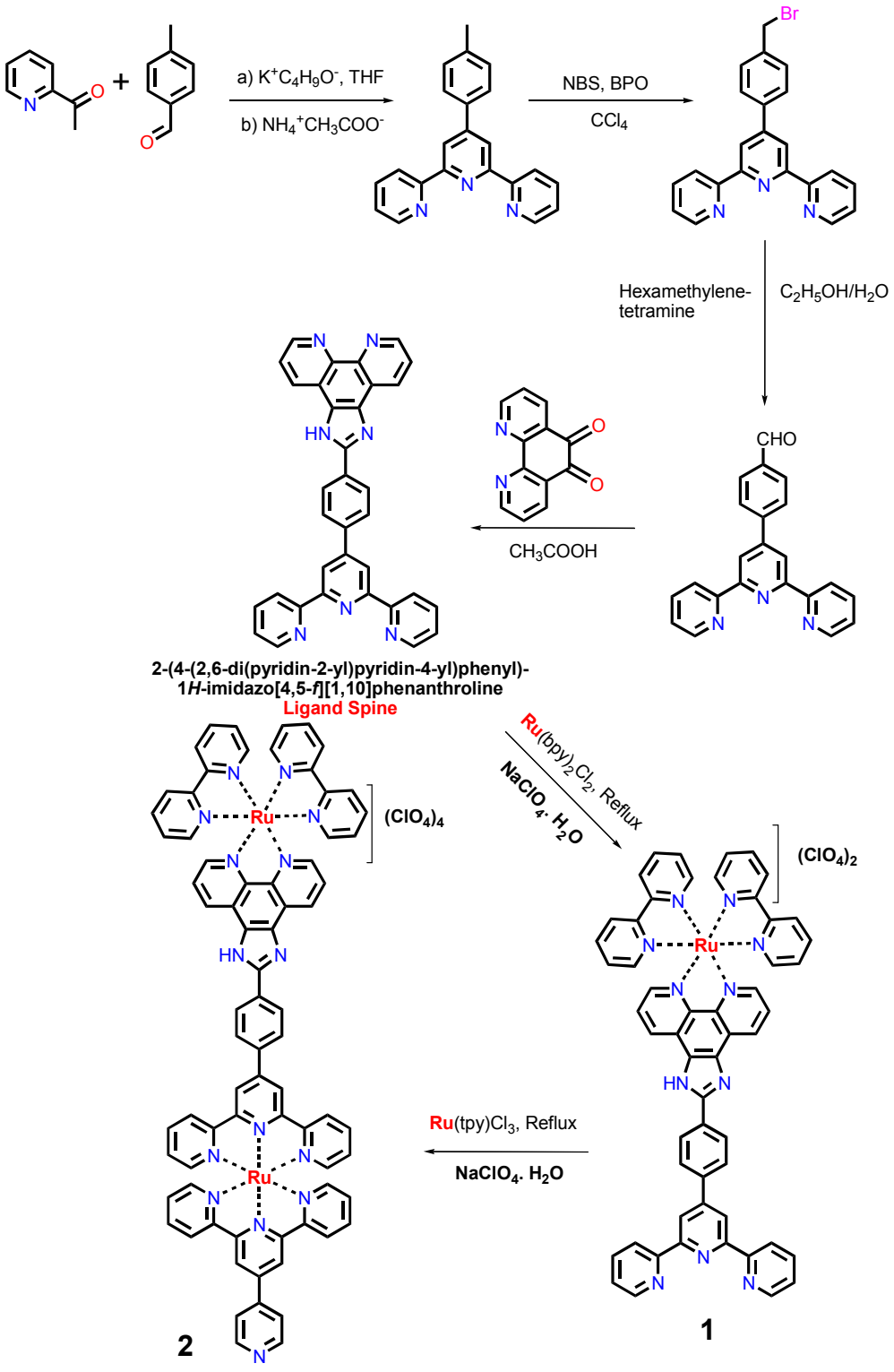

Scheme S1: Synthetic scheme for the homo-bimetallic chromophore **2**.

Characterization of **2**:

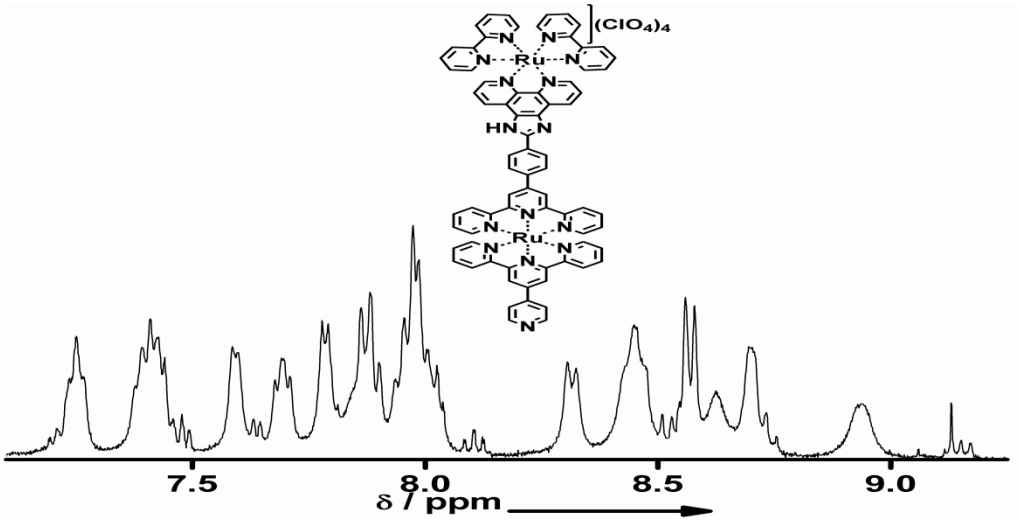

Figure S1: ^{1}H-NMR of **2** in DMSO-d_6 at ambient temperature.

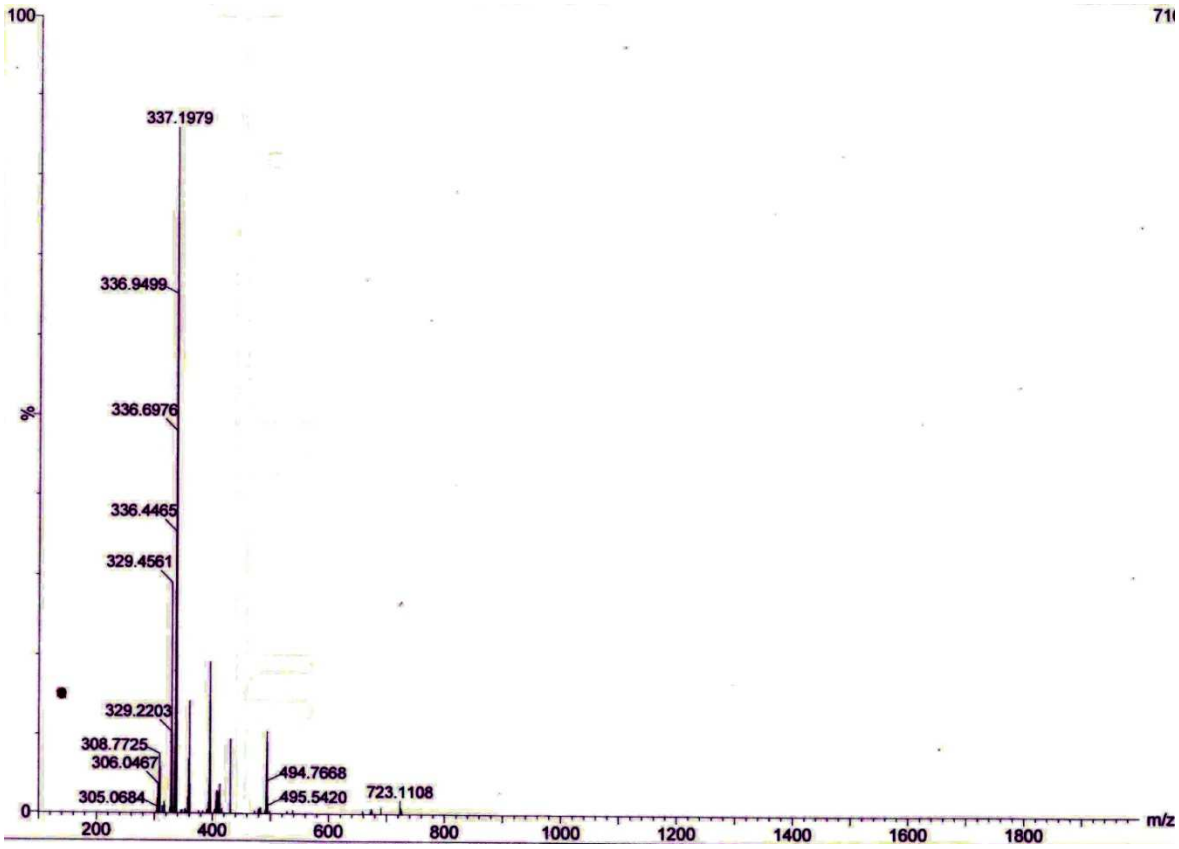

Figure S2: ESI-MS of **2** in acetonitrile at ambient temperature.

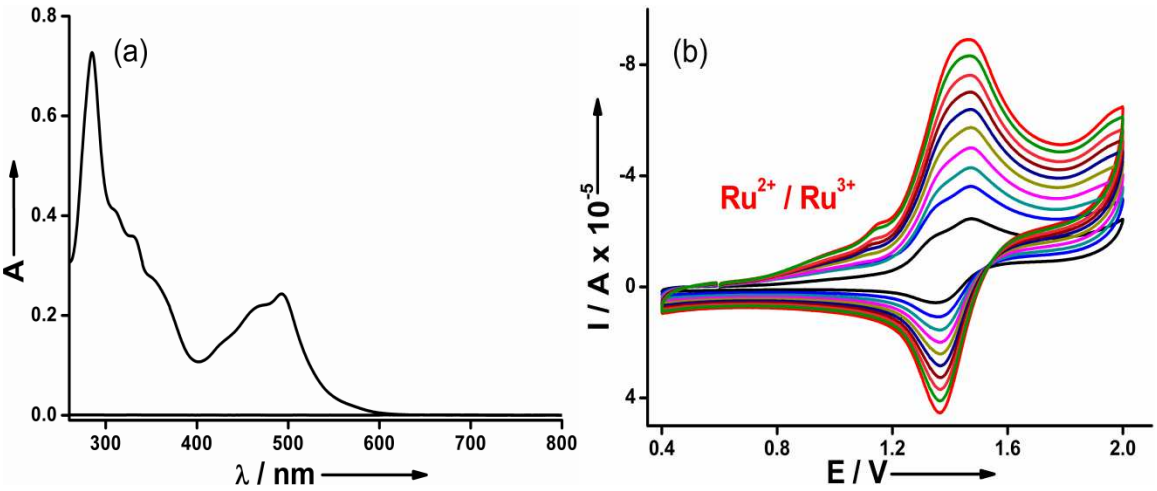

Figure S3: UV-vis spectrum of **2** (0.87 × 10^{-5} M, CH_3CN) at ambient temperature (a), Cyclic voltammetric responses of **2** (0.95 × 10^{-5} M, 10^{-3} M TBAP, CH_3CN) at different scan rates ranging from 100 - 1000 mVs^{-1}.

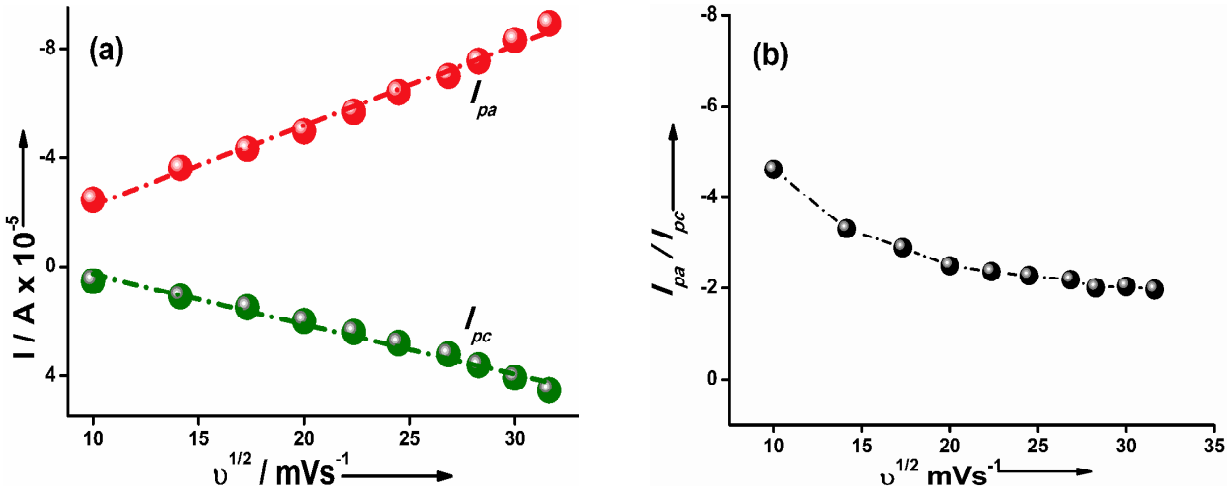

Figure S4: Plot showing linear fit of anodic (I_{pa}, red balls) and cathodic (I_{pc}, green balls) peak currents of **2** (0.95 × 10^{-5} M, 10^{-3} M TBAP, CH_3CN) as a function of scan rates (R^2 = ~ 0.98) (a), Plot of the peak current (I_{pa} / I_{pc}) ratio as a function of scan rates (b).

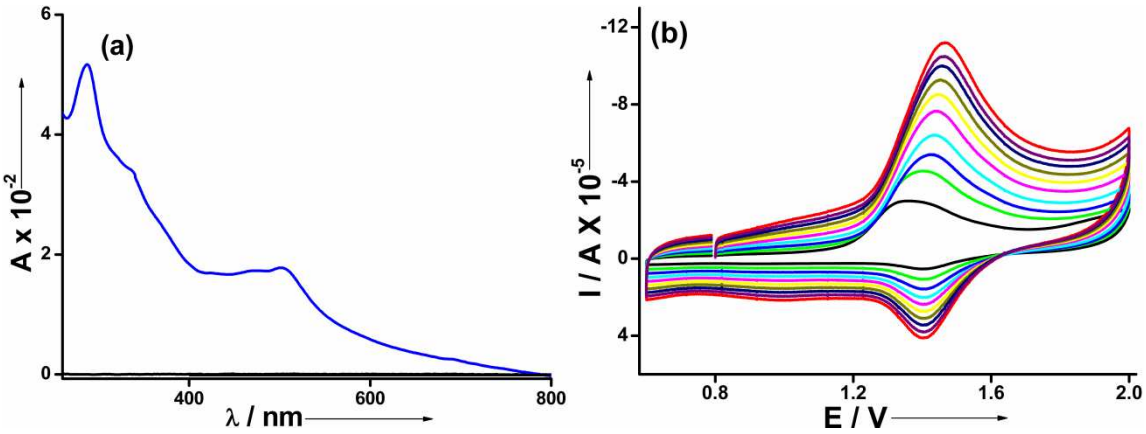

Figure S5: UV-vis spectrum of **2** on ITO monolayer (0.8 × 3 cm, CH_3CN) at ambient temperature (a), Cyclic voltammetric responses of **2** on ITO monolayer (0.8 × 3 cm, 20 mM TBAP, CH_3CN) at different scan rates ranging from 100-1000 mVs^{-1} (b).

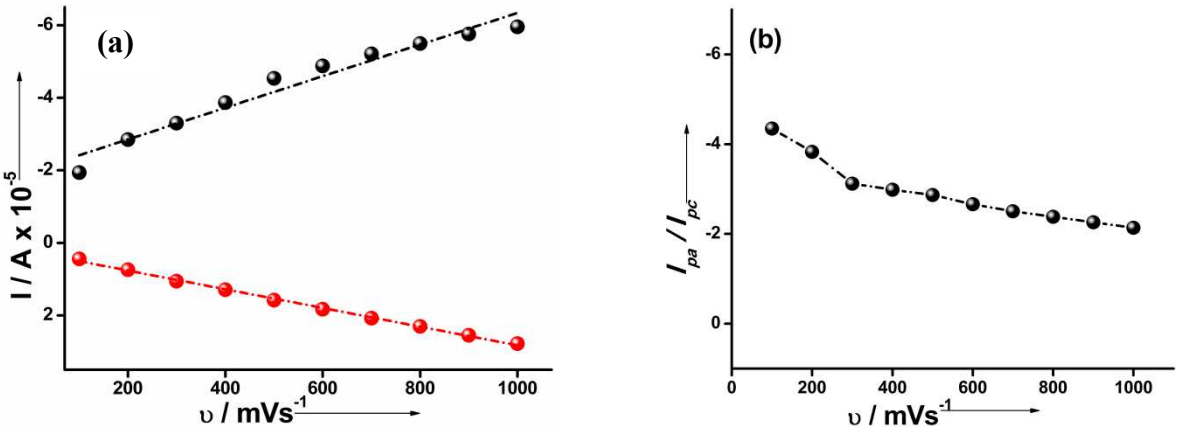

Figure S6: Plot showing the linear fit of anodic (I_{pa}, black balls) and cathodic (I_{pc}, red balls) peak currents of **2** on ITO monolayer (0.8 × 3 cm, 20 mM TBAP, CH_3CN) (R^2 = ~ 0.99) (a), as a function of scan rates (a). Plot of the peak current (I_{pa} / I_{pc}) ratio as a function of scan rates (b).

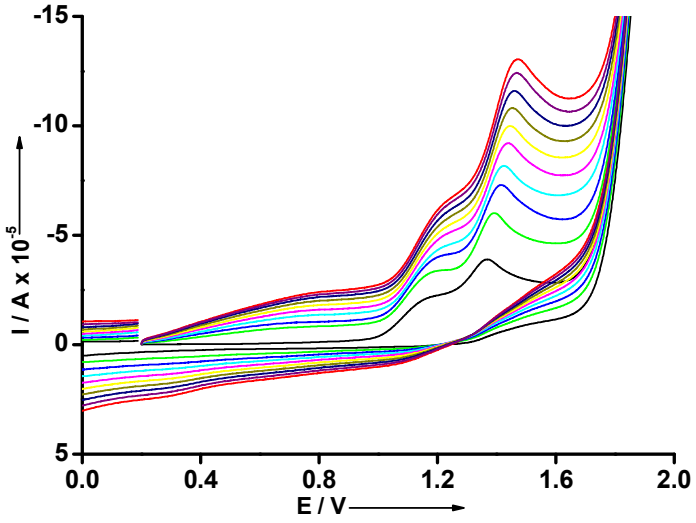

Figure S7: Cyclic voltammetric responses of ligand spine (0.87×10^{-5} M, 10^{-3} M TBAP, CH_3CN) at different scan rates ranging from 100-1000 mVs^{-1}.

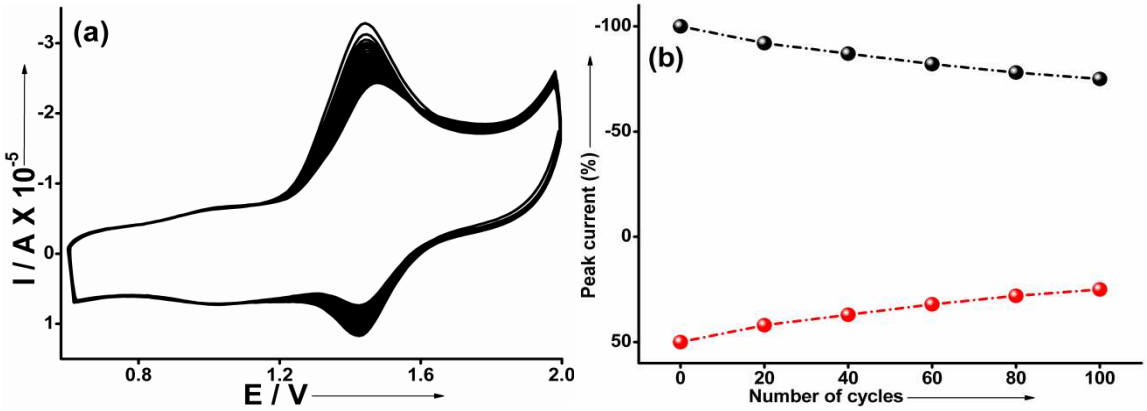

Figure S8: Read-write cycles of **2** on ITO for ruthenium metal centre (a). Anodic peak current (black balls) and cathodic peak current (red balls) as function of scan rate for 100 cycles (b).

Multi-potential switching of the absorbance of the 2 on ITO: The **2** on ITO was electro-optically switched by applying various potential biases for 1 s intervals, starting from 0.60 V and to 0.6 + *n* 0.1 V where *n* = 1, 2, 3…so on for 1 s. The monitoring the MLCT band at λ = 502 nm

would result in precise control over the transparency of monolayer, as ΔA is a function of the voltage until complete (100%) oxidation is achieved.

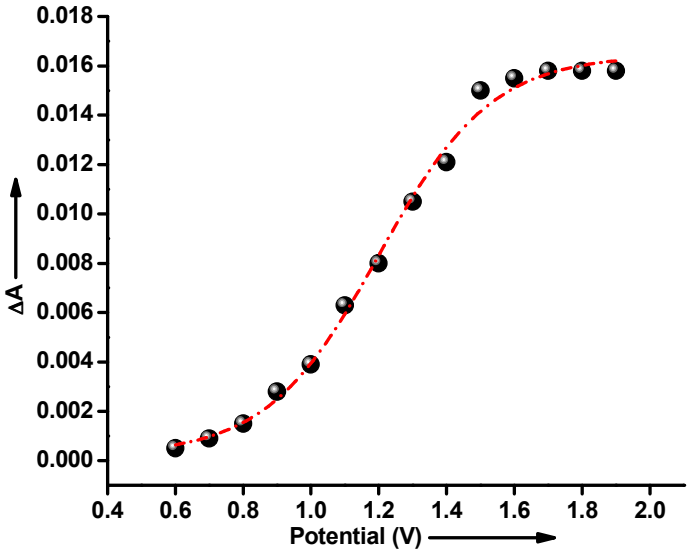

Figure S9. Difference in absorbance (ΔA) of the MLCT band at λ = 502 nm of **2** on ITO, as a function of the voltage, upon applying potential biases starting from 0.6 V to 0.6 + n 0.1 V where n = 1, 2, 3…so on for 1 s. Each data point represents the average of 3 measurements. The dashed red line is a sigmoidal fit (R^2 =0.98) of the data.

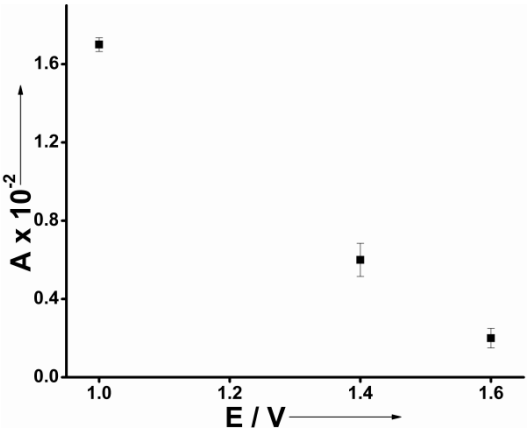

Figure S10: Details of triple-step potential based-switching carried out for three monolayers of **2** on ITO using the same set-up.

Electrochromic test in solution: Accumulative optical tuning of ruthenium. The electrolytic solution of homo-binuclear (0.97×10^{-5}M, 20 mM TBAP, 50 ml) was purged with N_2 in the bulk electrolysis kit for 15 min with mild stirring. Then, potential biases of 1.6 - 1.0 V and 1.0 - 1.4 - 1.6 V were applied for the redox tuning of the ruthenium centers and spectra were recorded as a function of time. The reversibility was tested for 3 successive cycles.

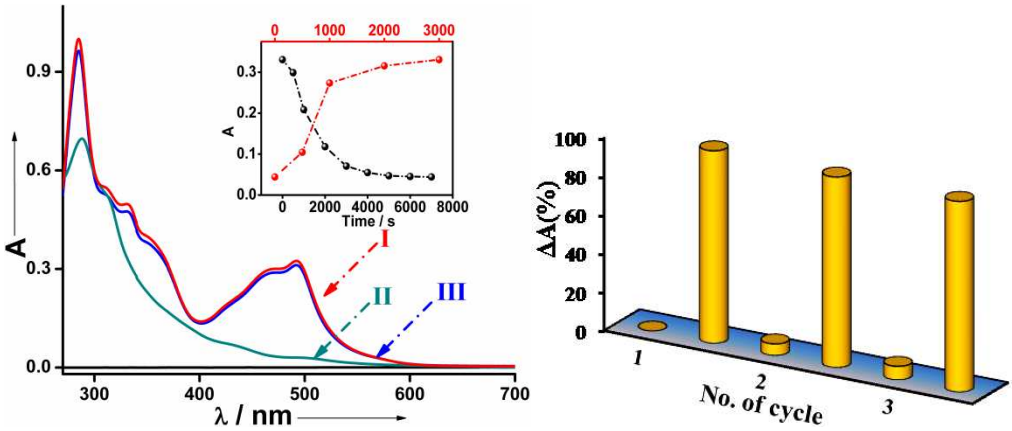

Figure S11: Absorbance intensity changes on electrochemical tuning of the ruthenium centers of the homo-binuclear complex (0.97×10^{-5} M, 20 mM TBAP in acetonitrile) in its initial spectrum (I) on applying 1.6 V (oxidation potential) (II), on applying 1.0 V (reduction potential) (III). Inset: representative plot for showing the gradual absorbance changes on applying the 1.6 V oxidation potential (black balls) and 1.0 V reduction potential (red balls), and bar chart for showing ΔA (%) at 492 nm (MLCT band position in solution study) *vs.* number of alternative redox cycles.

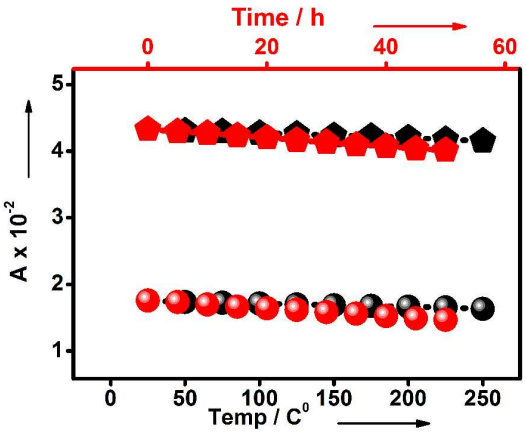

Figure S12: UV-vis monitoring of the **2** on ITO coated glass at various temperatures (25 – 250 °C) and time intervals (1 - 50 h) at 250 °C.

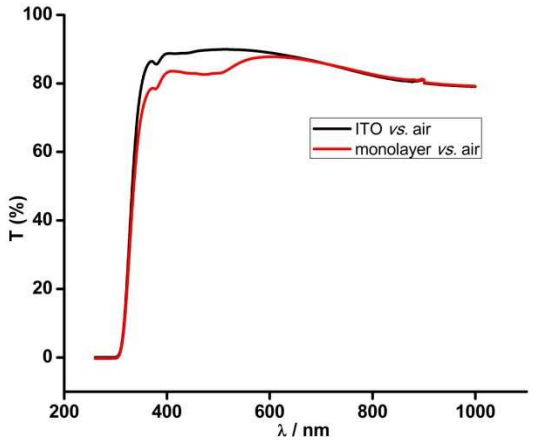

Figure S13: UV-vis transmittance spectra of ITO (black line) and **2** on ITO monolayer (0.8 × 3.0 cm).