Supporting Information Available

Cytochrome *c* Assembly on Fullerene Nanohybrid Metal Oxide Ultrathin film

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QCM Measurements.

A quartz crystal microbalance (QCM, 9 MHz) electrode manufactured by USI System, Japan, was used for monitoring film assembly and working electrode. The mass increase due to adsorption can be estimated form the QCM frequency shift by using Sauerbrey equation [50]. The following relationship is obtained between adsorbed mass, M(g), and frequency shift, ΔF (Hz), by taking into account the characteristics of used quartz resonators:

$$\Delta F (\text{Hz}) = -1.832 \times 10^{-8} \, M \,/\, A \tag{1}$$

, where A is the surface area of the resonator (0.159 cm²). In our system, a frequency shift of 1-Hz corresponds to a mass increase of ca. 0.9 ng. The thickness (d, Å) of an adsorbed film on one side of a resonator is given by

$$2d = \Delta F / 1.832\rho \tag{2}$$

, where ρ is the density (g/cm³) of the adsorbed film.



Figure S1. 2D AFM images of the 1 μ m × 1 μ m of the 1-cycle (a) TiO₂, (b) TiO₂/C₆₀, (c) TiO₂/C₆₀/TiO₂, and (d) (TiO₂/C₆₀/TiO₂)/Cyt. *c* layers deposited on a plasma treated silicon substrate, respectively.



Figure S2. Micrograph of surface contact angle of the outmost C_{60} layer deposited on a quartz palate.



Figure S3. Schematic illustration of the covalent bonding between C_{60} and TiO_2 gel layer.