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Unique Sequence-Dependent Properties of Trinucleotide Repeat Monolayers: Electrochemical, Electrical, and Topographic Characterization

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Tarlov's Method.

Number of $Ru(NH_3)_6^{3+}$ ion electrostatically binds with DNA equal to number of phosphate groups in the backbone. The binding of the ion enhances charge on the surface. The difference in charge in absence and presence of $Ru(NH_3)_6^{3+}$ can be obtained from chronocoulometry curve and using Cottrell equation (Eq. 1) the number of $Ru(NH_3)_6^{3+}$ can be calculated. The first term on right side of the Eq 1. is 'zero' because the charge due to diffusion (D_0) is 0 at t = 0, therefore the surface charge is the sum of double-layer charge (Q_{dl}) and the charge due to the cation ($nFA\Gamma_{Ru}$). Plugging the difference 'Q - Q_{dl}', number of electron transfer (n), Faraday constant (F), area of electrode (A) into Eq. 2, the number of $Ru(NH_3)_6^{3+}$ (Γ_{Ru}) can be calculated. Then, surface density of DNA, Γ_{DNA} can be calculated from the relationship Eq. 3 where z = charge of the cation, m = number of phosphate groups in each ds-DNA, and NA = Avogadro number.

$$Q = \frac{2nFAD_0^{1/2} C_0^*}{\pi^{1/2}} t^{1/2} + Q_{dl} + nFA\Gamma_{Ru} \qquad Eq.1$$

$$\Gamma_{Ru} = \frac{Q}{nFA} \qquad Eq.2$$

$$\Gamma_{DNA} = \Gamma_{Ru} \frac{z}{m} N_A \qquad Eq.3$$

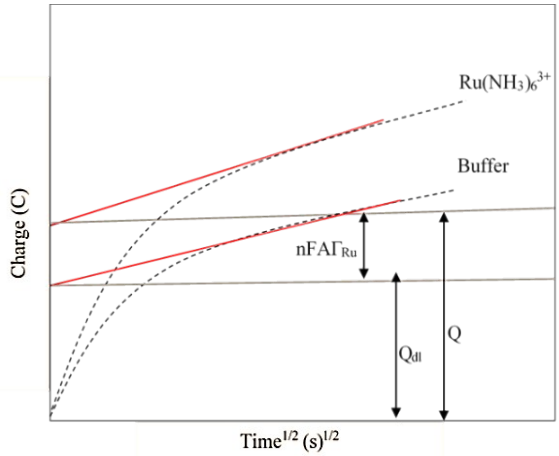

Figure S1. Typical chronocoulometric curves in absence and presence of $Ru(NH_3)_6^{3+}$.

Table S1. Surface density of double-stranded TNR sequences in number of strands, moles, and mass per centimeter square surface.

Sequence	Surface Density x10^{12} molecules/cm^2	Molecular Weight* g/mol	Density in Moles pmol/cm^2	Density in mass ng/cm^2
CGG-8	5.7	15011.24	9.46	142
CCG-8	2.9	15011.24	4.81	72.3
CTG-8	0.19	15003.24	0.32	4.73
CAG-8	0.33	15003.24	0.55	8.22
GAA-8	0.88	14995.24	1.46	21.9
GAA-5	1.83	9422.36	3.04	28.6
GAA-4	2.73	7567.74	4.53	34.3

*Molecular weight also includes the weight of tethering linker -$S(CH_2)_6OH$= 133.24 g/mol.

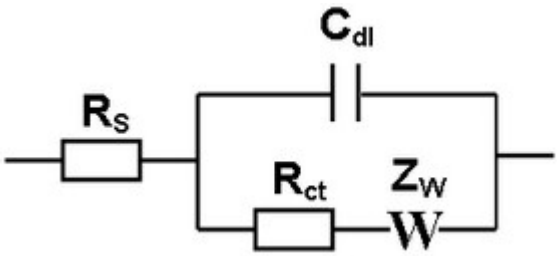

Figure S2. Randle's equivalent circuit used for fitting the experimental EIS data. R_s = solution resistance, Cdl = double layer capacitance, R_{ct} = charge transfer resistance, and W = Warburg impedance

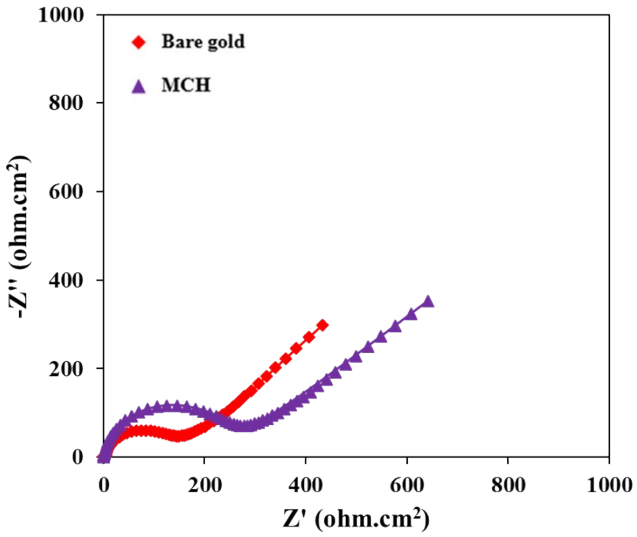

Figure S3. Nyquist form of EIS plots with curve fits for bare gold surface and mercaptahexanol (MCH) layer on gold electrode. The average value of MCH was measured to be 285 ± 162 Ω·cm^2 for N = 4 replicates.

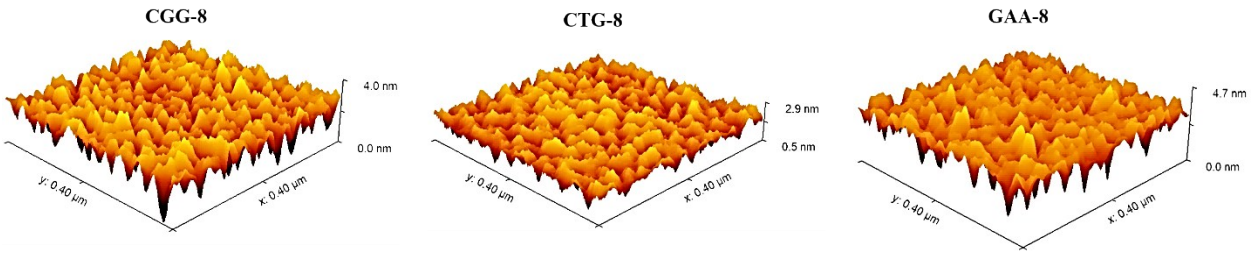

Figure S4. Tapping mode-AFM height images of (0.4 x 0.4 µm) ultra-flat gold surface, surface-tethered films of 10 µM dsCGG-8, dsCTG-8, and dsGAA-8 repeats on the gold substrate.