Redox-Triggered Hydrogels Revealing Switchable Stiffness Properties and Shape-Memory Functions

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Evaluation of the average molecular weight of the terpyridine-modified polyacrylamide chains.

Figure S1: (A) Calibration curves corresponding to the diffusion coefficients of a series of polymers of known average molecular weights: (a) Poly(acrylic acid) (MW ~ 35,000 Da) (b) Poly(acrylic acid), sodium salt (MW ~ 70,000 Da) (c) Poly(acrylic acid), sodium salt (MW ~ 85,000 Da) (d) Poly(acrylic acid) (MW ~ 345,000 Da) (e) Poly(acrylamide-co-acrylic acid) (MW ~ 520,000 Da). The diffusion coefficients of the respective polymers were derived by recording the DOSY spectrum of each of the polymers. (B) The DOSY NMR spectrum of the terpyridine-functionalized polyacrylamide polymer chains. The experimental logarithm of the diffusion coefficient, log(D) = -11, was used to deduce the molecular weight of the polymer (MW = 265,000 Da) using the calibration curve presented in (A).

(A)





Evaluation of the loading of the polyacrylamide chains with the terpyridine <u>ligands.</u>



Evaluation of the loading of the nucleic acid tethers on the <u>carboxymethylcellulose.</u>

Figure S3: (A) Absorbance spectra corresponding to the addition of variable amounts of carboxymethylcellulose (MW 250,000) to a fixed concentration of the nucleic acid (i), 500 nM: (a) 0 μ M (b) 0.1 μ M (c) 0.3 μ M (d) 0.5 μ M (e) 0.75 μ M (f) 1 μ M. (B) Calibration curve corresponding to absorbance ratio λ_{200} nm/ λ_{260} nm as a function of the molar ratio DNA: carboxymethylcellulose units. The circle marked with an arrow corresponds to the λ_{200} nm/ λ_{260} nm value of the polymer P_B. The calibration curve indicates a molar ratio of the nucleic acid and carboxymethylcellulose units that corresponds to 1:180.





(B)

SEM images of the Ru²⁺- and Ru³⁺-crosslinked carboxymethylcellulose matrices

Figure S4: SEM images corresponding to: (A) The Ru³⁺-crosslinked carboxymethyl cellulose hydrogel. (B) The Ru²⁺-crosslinked carboxymethyl cellulose hydrogel.

(A)



