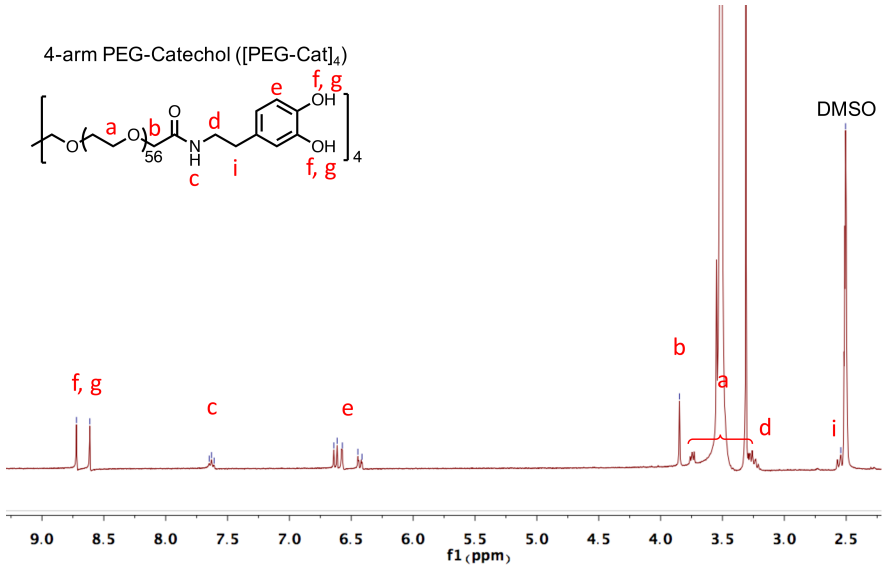

Figure S1. ^{1}H NMR spectrum of catechol-bearing 4-arm poly(ethylene glycol) ([PEG-Cat]$_4$).

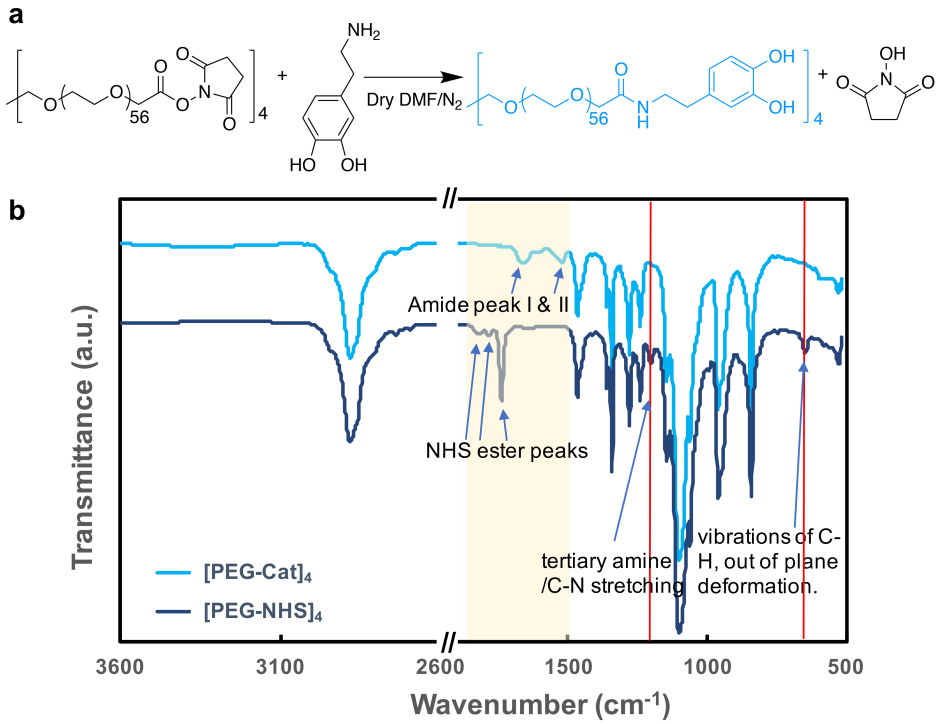

Figure S2. (a) Synthesis scheme of [PEG-Cat]$_4$. (b) FTIR spectra of [PEG-NHS]$_4$ and [PEG-Cat]$_4$.

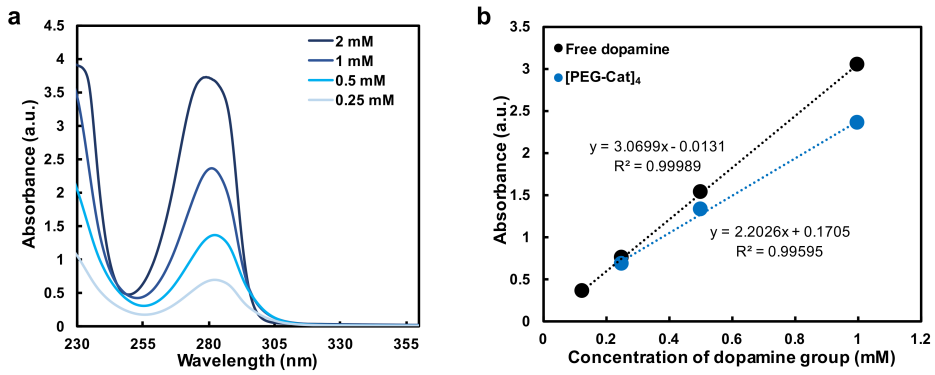

Figure S3. (a) UV-Vis spectra of solutions of 4-arm PEG-Catechol in water for select concentrations. (b) The calibration curve for catechol absorption at λ = 280 nm.

Table S1. Formulation of hydrogel precursors and associated redox reactions.

M^{z+}	Net Reaction[a]	$[M^{z+}]^d$ (mM)
Fe^{3+}	$2Fe^{3+} + QH_2 \rightarrow 2Fe^{2+} + Q + 2H^+$	64.0
VO_3^-	$2VO_3^- + 8H^+ + QH_2 \rightarrow 2VO^{2+} + Q + 4H_2O$	64.0
Ag^+	$2Ag^+ + QH_2 \rightarrow 2Ag + Q + 2H^+$	64.0
$AuCl_4^-$	$2[AuCl_4^-] + 3QH_2 \rightarrow 2Au + 3Q + 6HCl + 2Cl^-$	21.3
Cu^{2+} [b]	$2\ Cu^{2+} + QH_2 \rightarrow 2Cu^+ + Q + 2H^+$	64.0
Cu^{2+} [c]	$Cu^{2+} + QH_2 \rightarrow Cu + Q + 2H^+$	32.0
Al^{3+}	$2Al^{3+} + 3QH_2 \rightarrow 2Al + 3Q + 6H^+$	21.3
VO^{2+}	$2VO^{2+} + 2H^+ + QH_2 \rightarrow 2V^{3+} + 2H_2O + Q$	64.0

[a]Note: QH_2 – fully reduced catechol; Q – fully oxidized *o*-quinone.
[b]Reduction of Cu^{2+} to Cu^+
[c]Reduction of Cu^{2+} to Cu^0
[d]Concentration of $[PEG\text{-}Cat]_4$ held constant at 8 mM for all reactions.

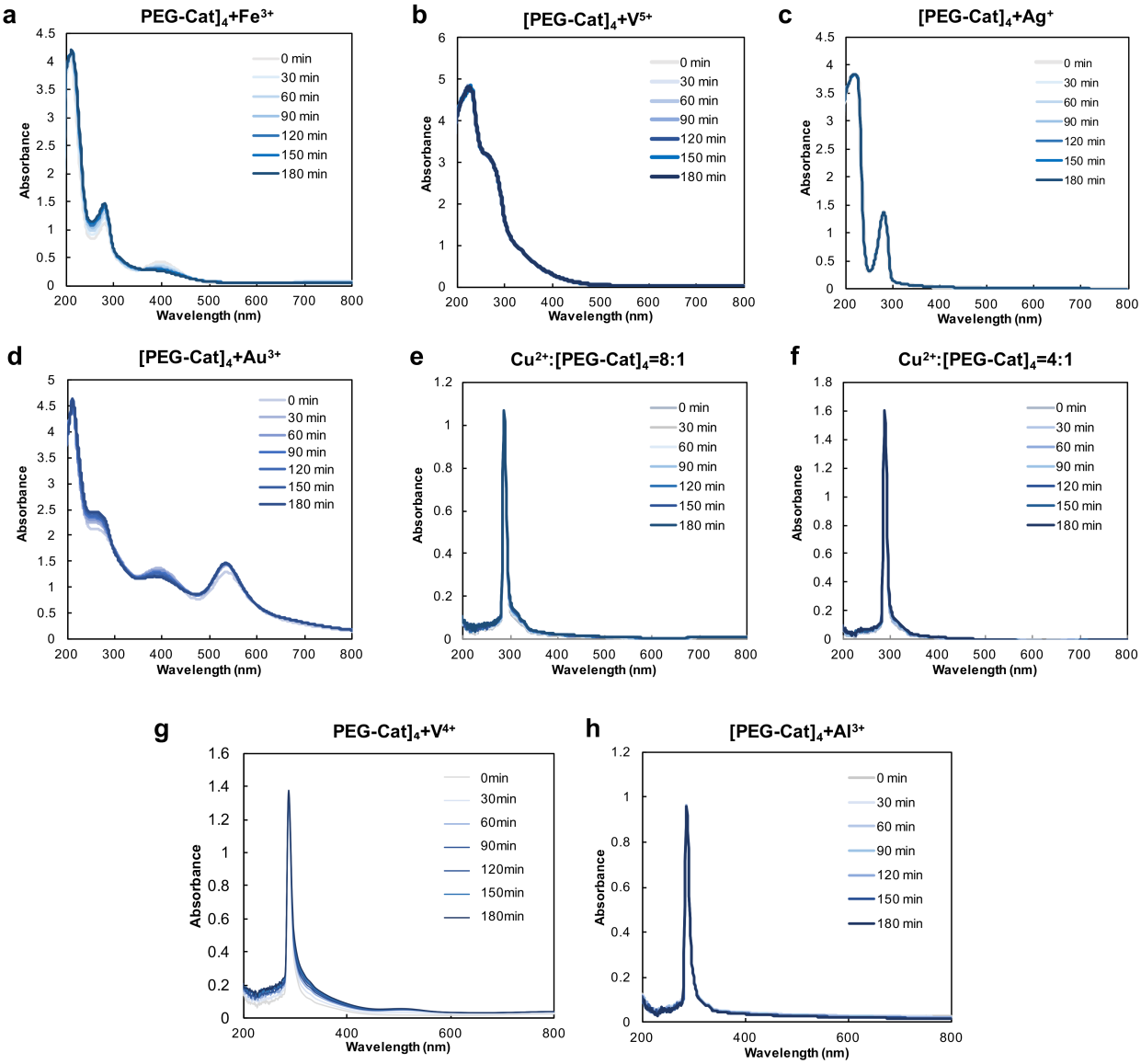

Figure S4. UV-vis spectra of dilute solutions of precursors as a function with time for the following compositions: (a) $[PEG\text{-}Cat]_4+Fe^{3+}$; (b) $[PEG\text{-}Cat]_4+V^{5+}$; (c) $[PEG\text{-}Cat]_4+Ag^{+}$; (d) $[PEG\text{-}Cat]_4+Au^{3+}$; (e,f) $[PEG\text{-}Cat]_4+Cu^{2+}$; (g) $[PEG\text{-}Cat]_4+V^{4+}$; (h) $[PEG\text{-}Cat]_4+Al^{3+}$.

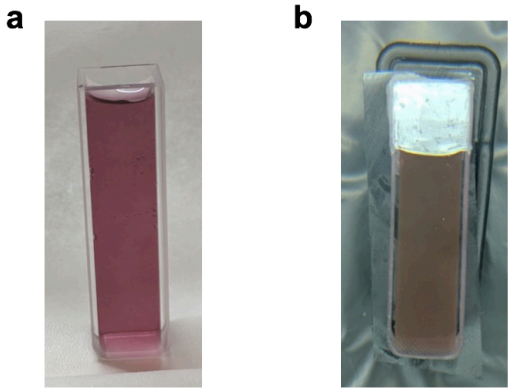

Figure S5. $[PEG\text{-}Cat]_4+Au^{3+}$ samples prepared for UV-vis spectroscopy imaged under (a) transmitted and (b) reflective light.

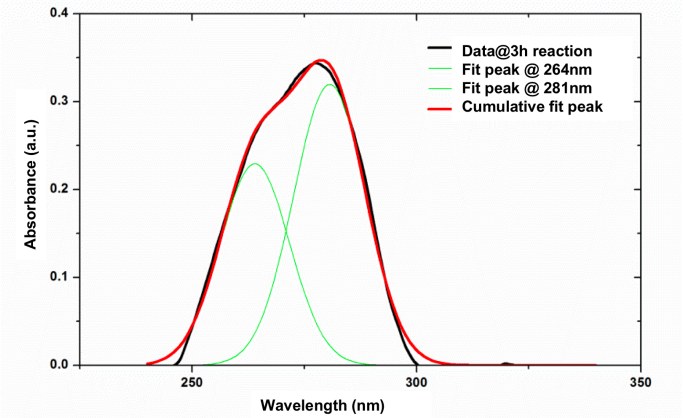

Figure S6. Peak deconvolution and fitting with baseline correction with UV-vis spectrum of $[PEG\text{-}Cat]_4+Au^{3+}$.