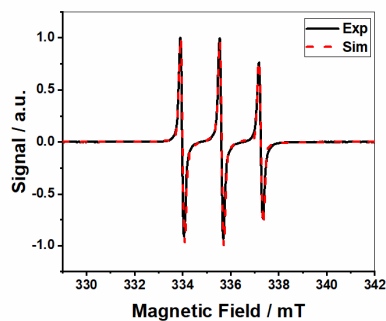
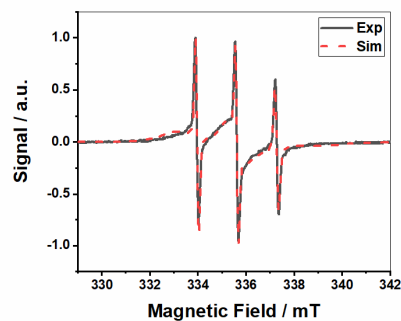


Figure S1. Time-dependent elastic ( $G'$ ) and viscous moduli ( $G''$ ) of 1000  $\mu$ L 3 mM BSA<sub>E</sub>(37, 7) with different amounts of ethanol.

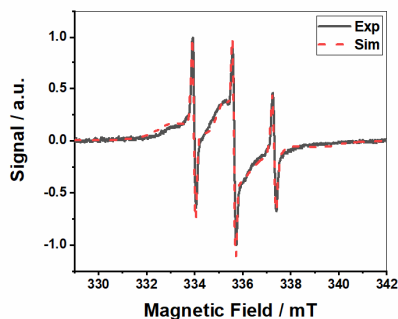
(A) 120  $\mu\text{L}$  5 mM BSA<sub>E</sub>(37, 7, 2) + 160  $\mu\text{L}$  EtOH + 1.2:1 SL-NPX:BSA



(B) 120  $\mu\text{L}$  5 mM BSA<sub>E</sub>(37, 7, 4) + 80  $\mu\text{L}$  EtOH + 1.2:1 SL-NPX:BSA



(C) 120  $\mu\text{L}$  5 mM BSA<sub>E</sub>(37, 7, 24) + 80  $\mu\text{L}$  EtOH + 1.2:1 SL-NPX:BSA



(D) 120  $\mu\text{L}$  5 mM BSA<sub>E</sub>(37, 7, 24) + 80  $\mu\text{L}$  EtOH + 2.4:1 SL-NPX:BSA

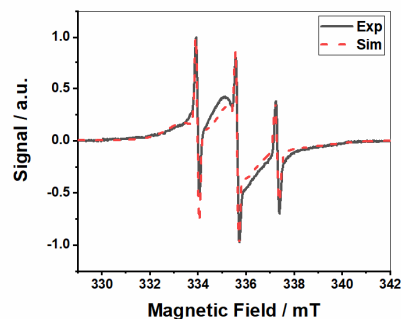


Figure S2. (A) SL-NPX loaded BSA<sub>E</sub>(37, 7, 2) at a 1.2:1 SL-NPX: BSA molar ratio with 160  $\mu\text{L}$  ethanol, (B) SL-NPX loaded BSA<sub>E</sub>(37, 7, 4) at a 1.2:1 SL-NPX: BSA molar ratio with 80  $\mu\text{L}$  ethanol, (C) SL-NPX loaded BSA<sub>E</sub>(37, 7, 24) at a 1.2:1 SL-NPX: BSA molar ratio with 80  $\mu\text{L}$  ethanol and (D) SL-NPX loaded BSA<sub>E</sub>(37, 7, 24) at a 2.4:1 SL-NPX: BSA molar ratio with 80  $\mu\text{L}$  ethanol.

Table S1. Parameters of spectral simulation of Figure S2.

Figure	Type of components	Percentage	Correlation time $\tau_c$ [ns]	Hyperfine coupling constant $a_{\text{iso}}$ [MHz]
A	Bound	0	0	0
	Intermediate	51%	2	44.5
	Free	48%	0.11	45.8
B	Bound	0	14	46.1
	Intermediate	94%	9	47.5
	Free	6%	0.1	46.5
C	Bound	2%	13	47.1
	Intermediate	95%	4	46.2
	Free	3%	0.47	46.8
D	Bound	5%	15	44.7
	Intermediate	91%	8	46.5
	Free	3%	0.46	46.5

By comparing the parameters obtained from EPR spectral simulation of figure S2A with S2B we can realize that increasing the amount of ethanol leads to lower percentage of freely tumbling SL-NPX. Moreover, increasing the incubation time from 4 hours to 24 hours (see Figure S2C and S2D) results in the higher percentage of strongly bound SL-NPX to BSA in the hydrogel.

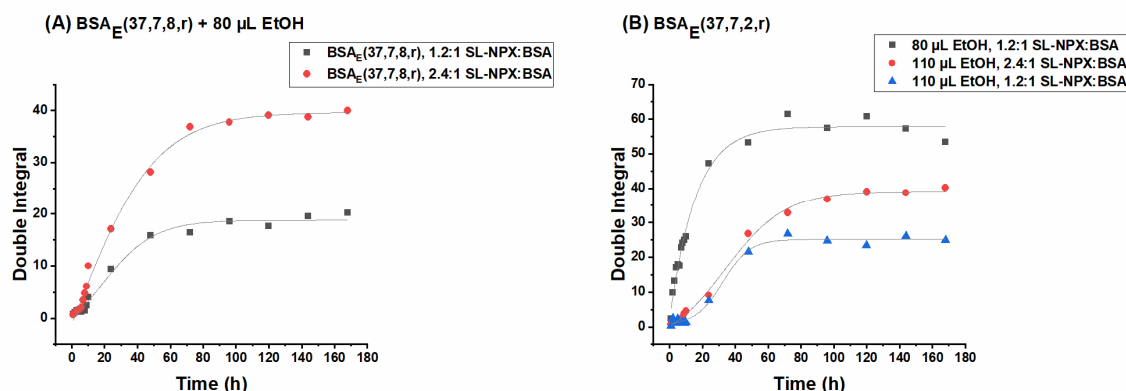


Figure S3. Double integral values as a function of release time intervals for (A) SL-NPX loaded BSA<sub>E</sub>(37, 7, 8, r) prepared by addition of 80  $\mu$ L ethanol and different molar ratios of SL-NPX: BSA and (B) BSA<sub>E</sub>(37, 7, 2, r) prepared by addition of different ethanol amounts and different SL-NPX: BSA molar ratio. All of the samples are prepared with 120  $\mu$ L BSA 5 mM.

Figure S3A shows the direct effect of initial drug loading on release rate, as higher molar ratios of SL-NPX can lead to larger double integral value, i.e. higher release rate. As can be seen in Figure S3B, increasing the amount of ethanol from 80  $\mu$ L to 110  $\mu$ L results in lower release rate due to stronger mechanical properties. In rheological characterizations section, we have completely explain the effect of ethanol on viscoelastic behavior of the BSA hydrogels.

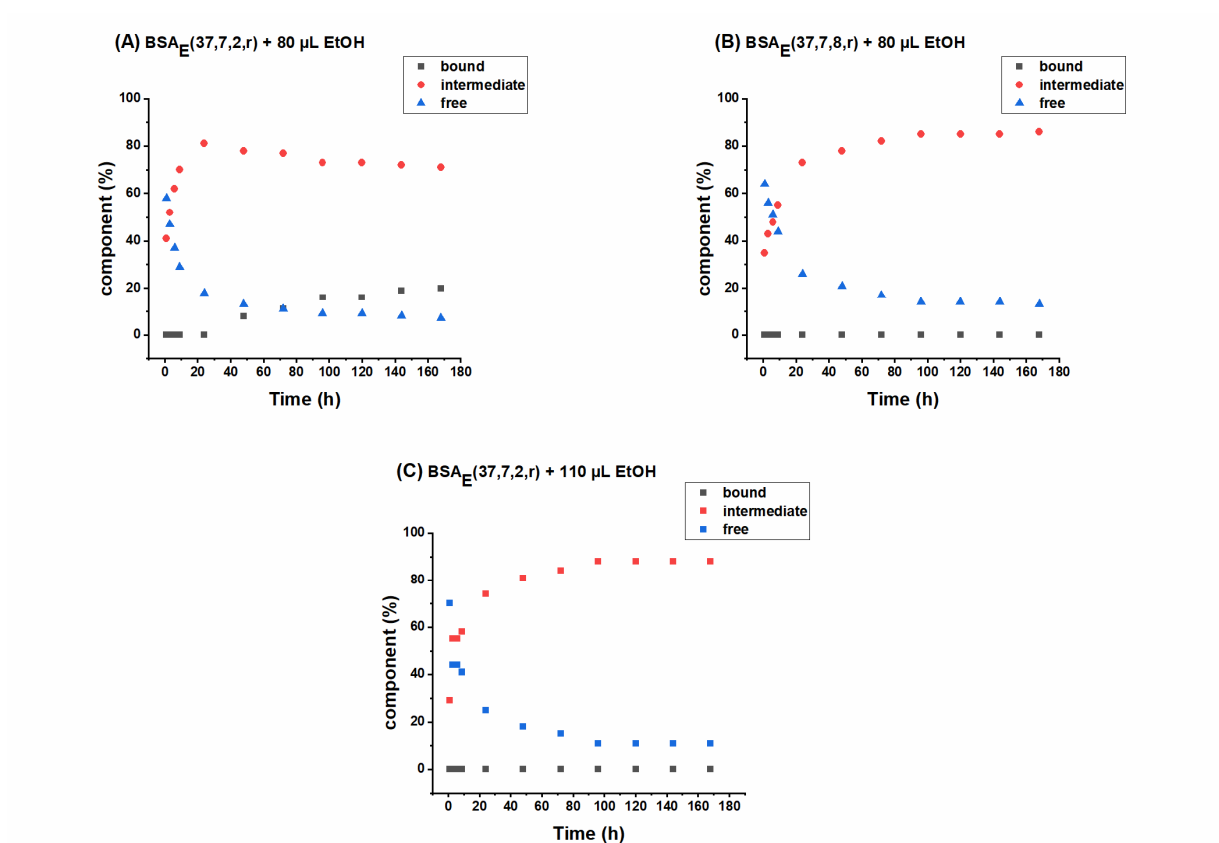


Figure S4. The released percentage of freely rotating SL-NPX, intermediately and tightly bound SL-NPX to BSA versus time intervals of (A)  $BSA_E(37, 7, 2, r)$  with  $80 \mu L$  ethanol at a 2.4:1 SL-NPX: BSA molar ratio, (B)  $BSA_E(37, 7, 8, r)$  with  $80 \mu L$  ethanol at a 2.4:1 SL-NPX: BSA molar ratio and (C)  $BSA_E(37, 7, 2, r)$  with  $110 \mu L$  ethanol at a 1.2:1 SL-NPX: BSA molar ratio. In these samples the precursor solutions are prepared with addition of  $200 \mu L$  BSA 5 mM.

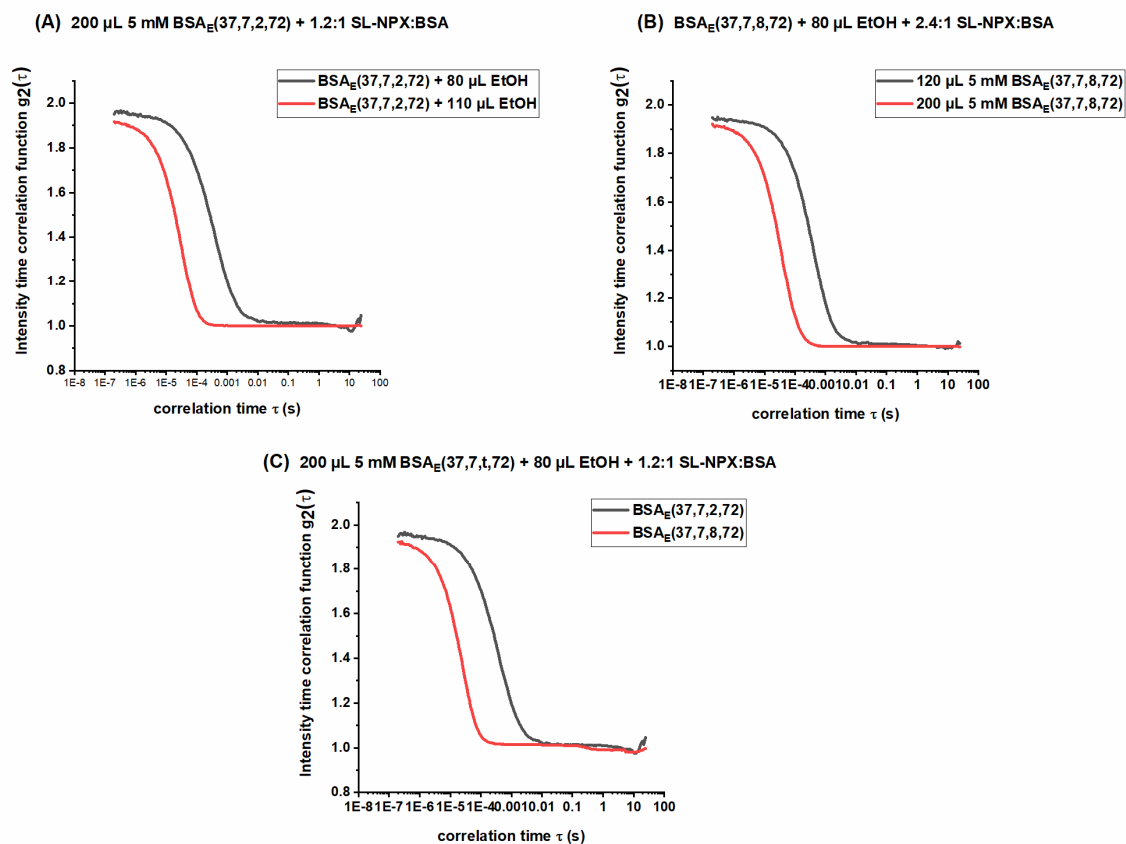


Figure S5. Intensity time correlation functions for (A) SL-NPX loaded  $\text{BSA}_E(37, 7, 2, 72)$  at a 1.2:1 SL-NPX: BSA molar ratio prepared with different amounts of ethanol, (B) SL-NPX loaded  $\text{BSA}_E(37, 7, 8, 72)$  at a 2.4:1 SL-NPX: BSA molar ratio prepared with 80  $\mu\text{L}$  ethanol and different BSA concentrations and (C) SL-NPX loaded  $\text{BSA}_E(37, 7, t, 72)$  at a 1.2:1 SL-NPX: BSA molar ratio prepared with 80  $\mu\text{L}$  ethanol.