SUPPLEMENTARY INFORMATION

Surfactant-Induced Assembly of Enzymatically-Stable Peptide Hydrogels

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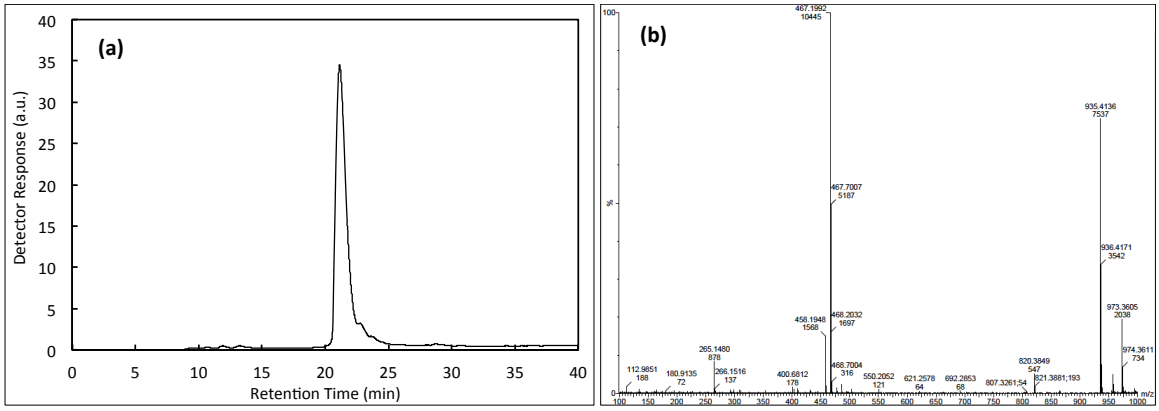

Figure S1. (a) HPLC trace of as-synthesized Bola-1. The trace shows a single peak at a retention time of 21 min, indicating the successful synthesis of highly pure Bola-1. **(b)** Mass spectrum of as-synthesized Bola-1. The spectrum shows peaks at m/z = 467.2 and m/z = 935.4, consistent with a doubly charged ion of m = $[M-2H]^{-2}$ and a singly charged ion of m = $[M-H]^{-}$, respectively, where M = 936.4 Da for Bola-1.

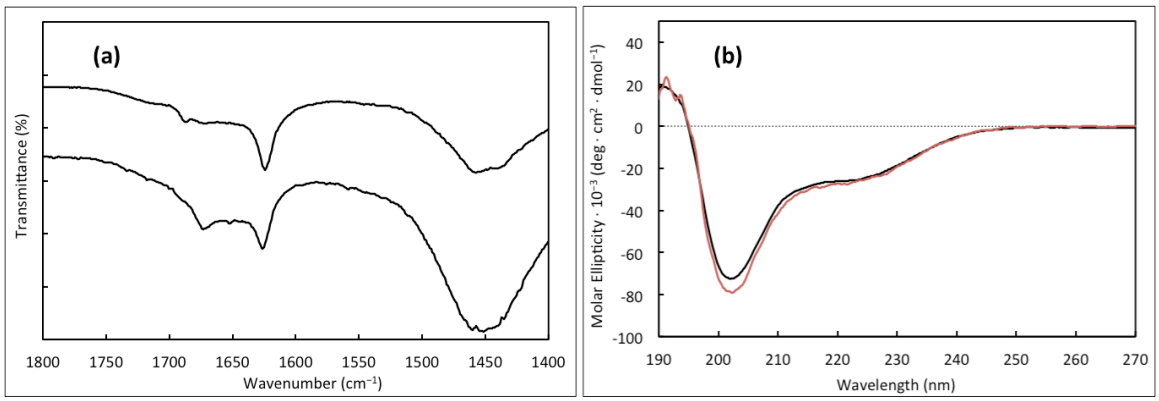

Figure S2. (a) FTIR and **(b)** CD spectra showing reproducibility by different sample preparation methods. In **(a)**, both spectra show 10 mM Bola-1 in D_2O to which 15 mM SDS is added to induce gelation. For the upper spectrum, the gel was fabricated directly on the ATR cell of the FTIR instrument, while for the lower spectrum, the gel was fabricated separately and then loaded on the ATR cell. The spectra are shifted vertically for clarity. In **(b)**, the black curve shows 0.2 mM Bola-1 directly added to 15 mM SDS. The red curve shows 10 mM Bola-1 in H_2O to which 15 mM SDS is added to induce gelation. A portion of this gel is then diluted with 15 mM SDS to 0.2 mM Bola-1.

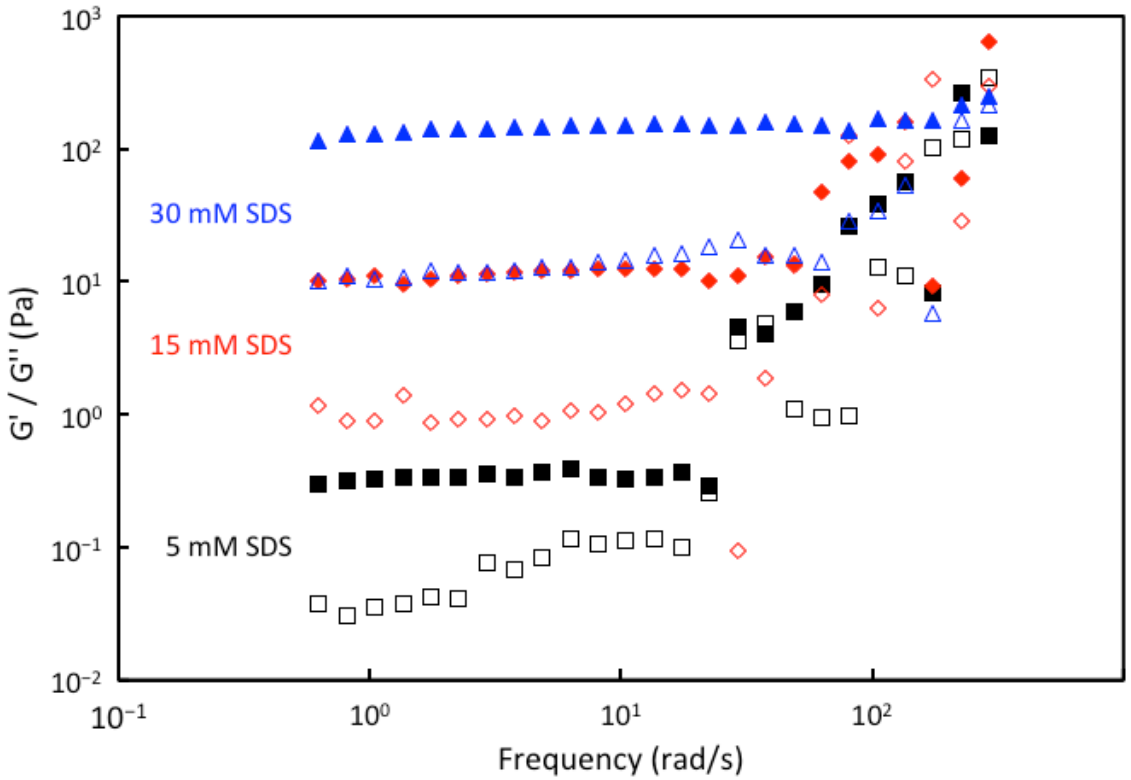

Figure S3. Dynamic frequency sweep data (strain = 1%) from Bola-1/SDS materials. The peptide concentration for all samples is 10 mM. The SDS concentration is marked to the left of the corresponding data sets. Filled and open symbols show the storage modulus, G', and the loss modulus, G'', respectively. In all three data sets, solid-like behavior ($G' > G''$) with essentially constant G' is exhibited at low frequency, persisting through the lowest frequency measured. This behavior is additional evidence of the formation of a crosslinked network and concomitant gelation induced in Bola-1 solutions by the addition of SDS. The scatter at higher frequencies is due to torque values below the sensitivity of the instrument, and may potentially reflect a breakdown of network structure at larger shear rates.

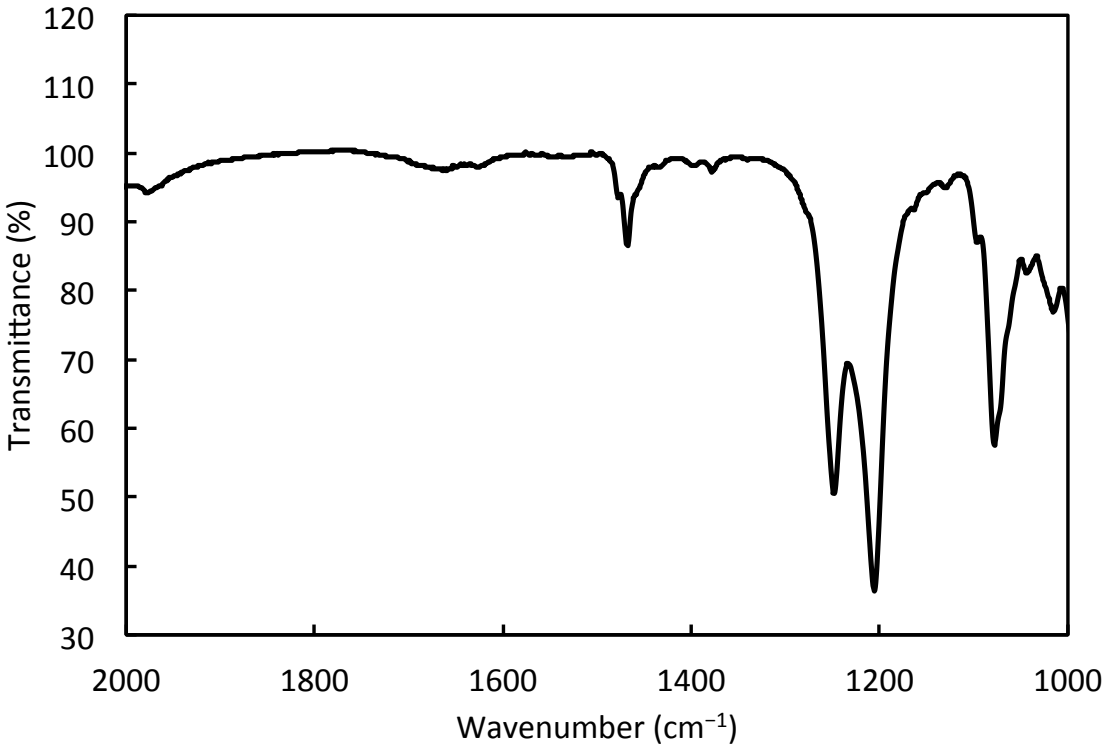

Figure S4. FTIR spectrum of pure SDS. SDS exhibits negligible absorption over the wavenumber range 1600-1700 cm^{-1}. Therefore, FTIR bands over this range in Bola-1/SDS materials can be ascribed to the amide I vibration of the peptide.

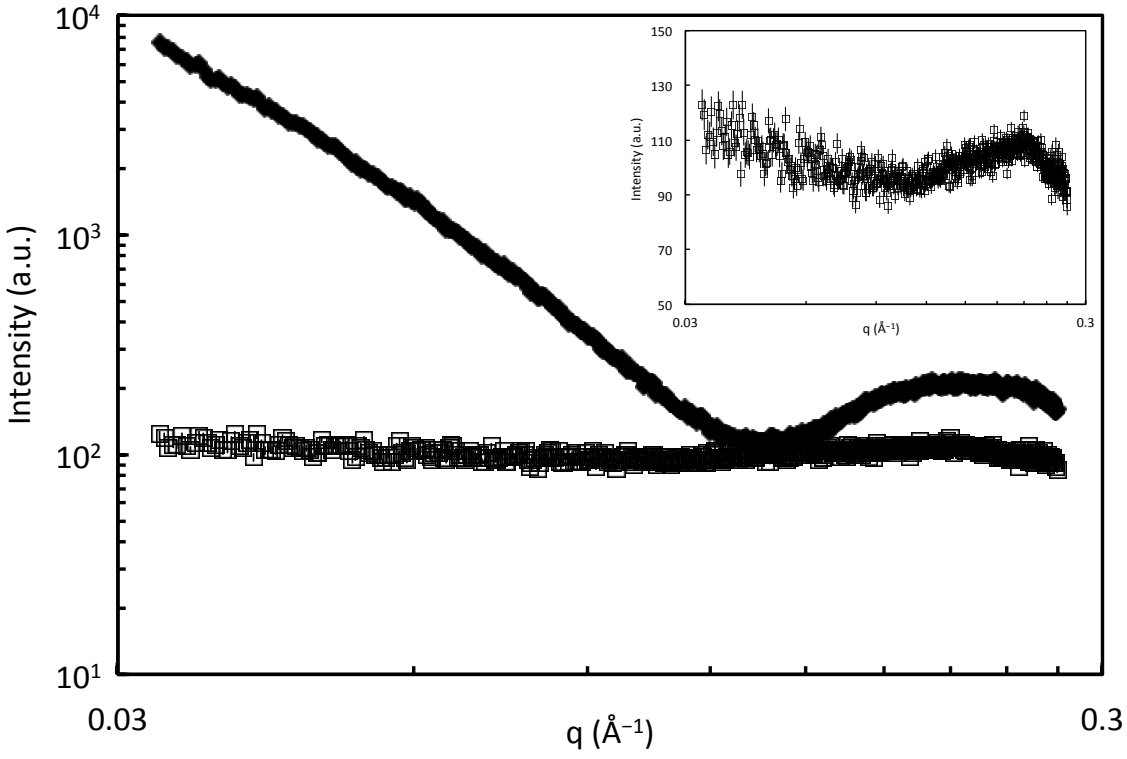

Figure S5. SAXS profiles of (◆) 10 mM Bola-1 with 15 mM SDS and (□) 15 mM SDS. No background subtraction has been performed on either profile. The inset shows the profile of 15 mM SDS plotted in a linear-log format. The error bars indicate ± one standard deviation. While the inset clearly shows the characteristic scattering pattern that is well established for SDS micelles, the relative contrast between the micelles and their surrounding medium is weak. The main panel illustrates the dominance of coherent scattering from a Bola-1/SDS gel over that from SDS micelles alone. Therefore, the observed *q*-dependent scattering from the Bola-1/SDS gel must arise exclusively from the ordered structure created through peptide-surfactant interactions; that is, the observed scattering is not simply that of standard SDS micelles.

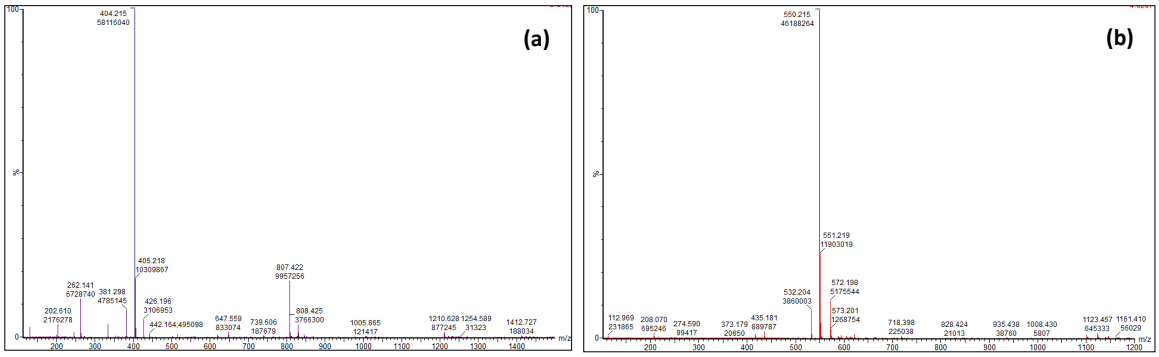

Figure S6. MS spectra of **(a)** 8 min elution product and **(b)** 23 min elution product of HPLC separation performed on 10 mM Bola-1 in H_2O with 80 µM CT (see Figure 6a and the accompanying discussion). Separation was performed and spectra were collected after one week's incubation of the Bola-1/CT solution at room temperature. The spectra in **(a)** and **(b)** show peaks at m/z = 404.2 and m/z = 550.2, respectively, consistent with the singly charged ions of m = $[M–H]^-$ for H-$(Ala)_2$-Lys-Asp-OH (M = 405.2 Da) and H-Asp-Glu-$(Ala)_2$-Phe-OH (M= 551.2 Da), which are the expected degradation products resulting from hydrolytic cleavage of Bola-1 at the C-terminus of the phenylalanine residue.

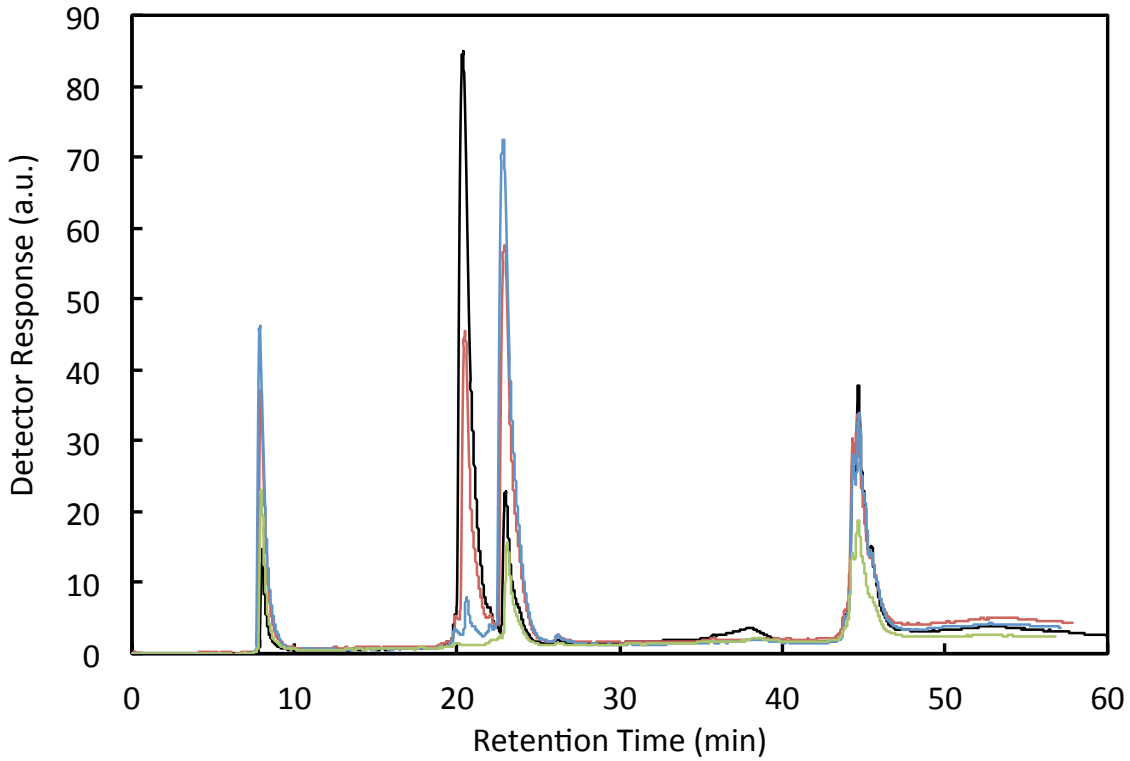

Figure S7. HPLC traces of 10 mM Bola-1 in H_2O with 80 μM CT (pH ~ 3) after incubation at room temperature for **(black)** 3 hr, **(red)** 1 day, **(blue)** 3 days, and **(green)** 6 days. The HPLC trace after 3 hr clearly shows detectable levels of the two cleavage products, which elute at 8 min and 23 min, along with uncleaved Bola-1, which elutes at 21 min. Furthermore, the traces show that, at this pH and concentration, the cleavage of Bola-1 by CT requires between 3 and 6 days for completion, as evidenced by the disappearance of the peak at 21 min.

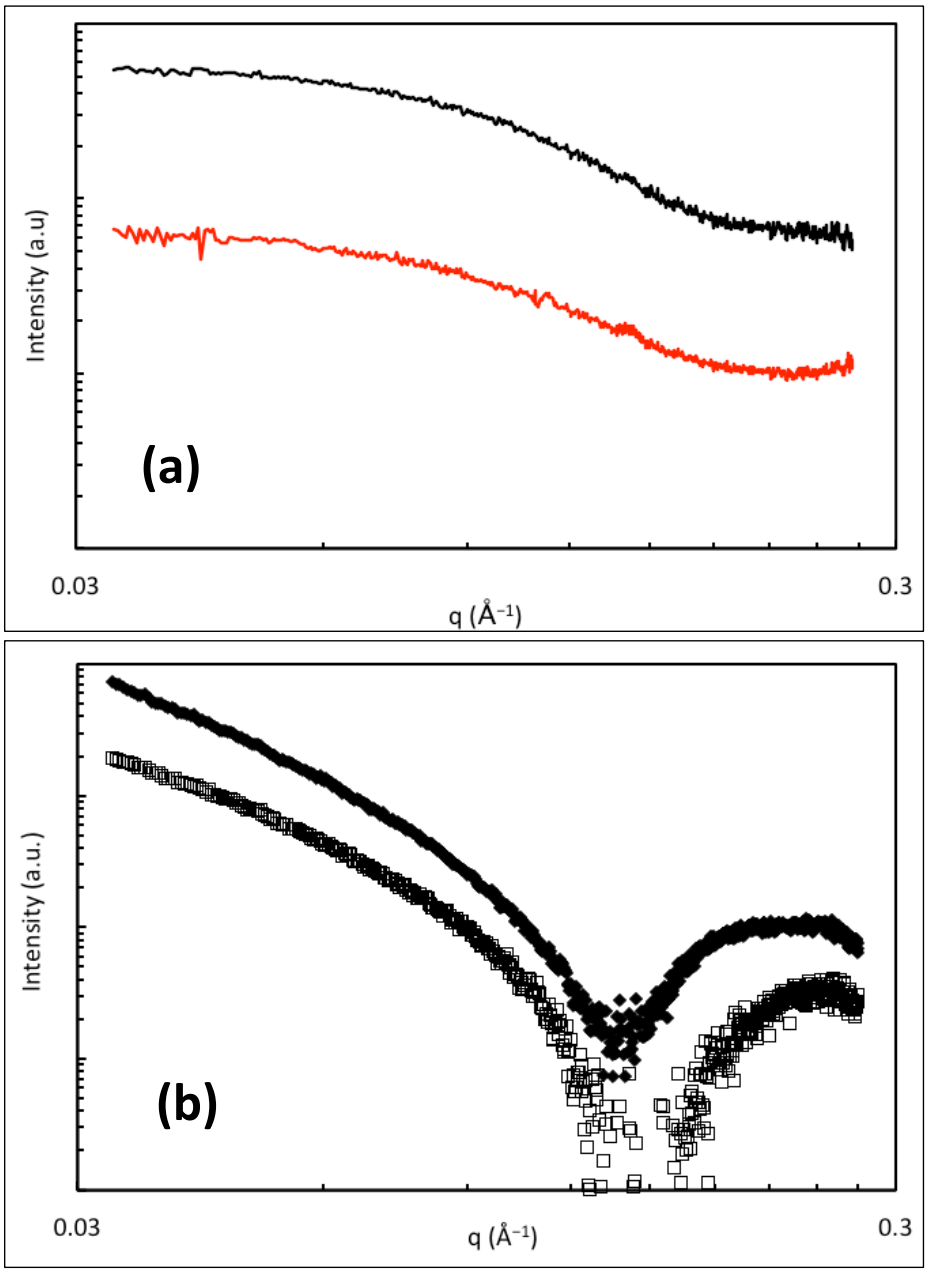

Figure S8. SAXS profiles of **(a)** chymotrypsin (80 μM) in water (red) and Bola-1 (10 mM) with chymotrypsin (80 μM) in water (black). In **(b)** profiles for 10mM Bola-1 + 15 mM SDS (◆**)** and 10 mM Bola-1 + 15 mM SDS + chymotrypsin (□) are effectively identical (small deviation in curves is due to scattering from the chymotrypsin (see S8a). Again the curves are shifted slightly for visual clarity.

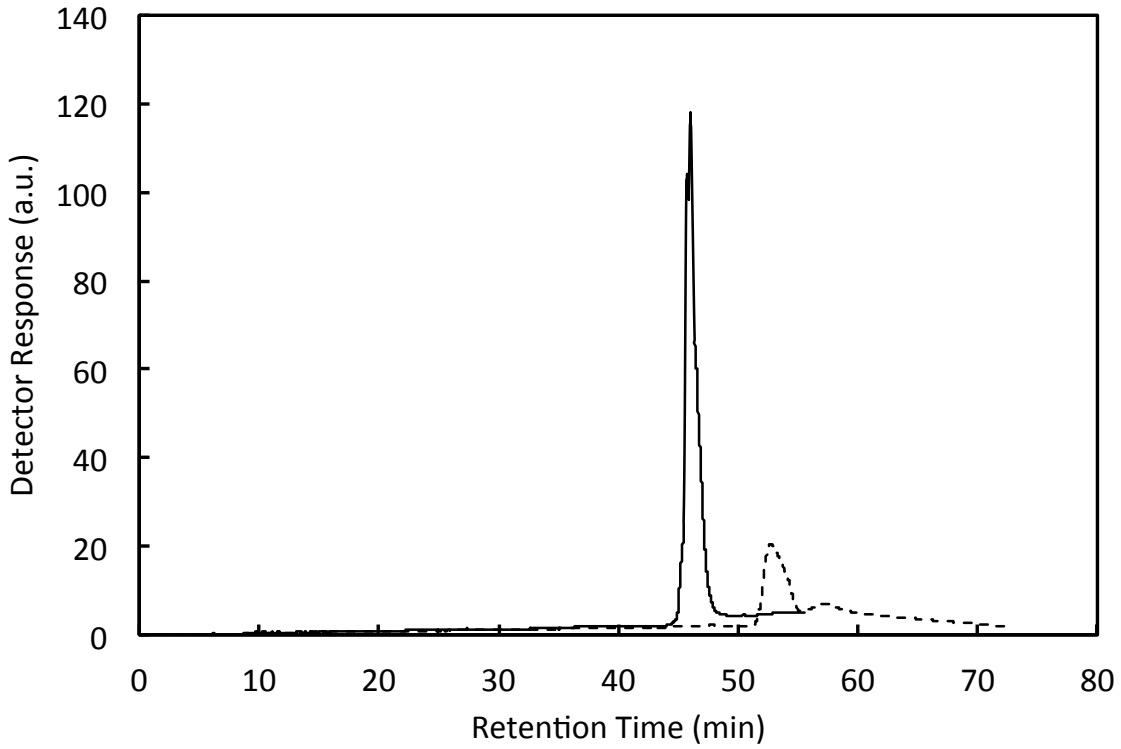

Figure S9. HPLC traces of **(solid)** CT and **(dashed)** 80 μM CT exposed to 15 mM SDS. The change in peak position and shape reflects the denaturation of CT by SDS.