Supporting Information:

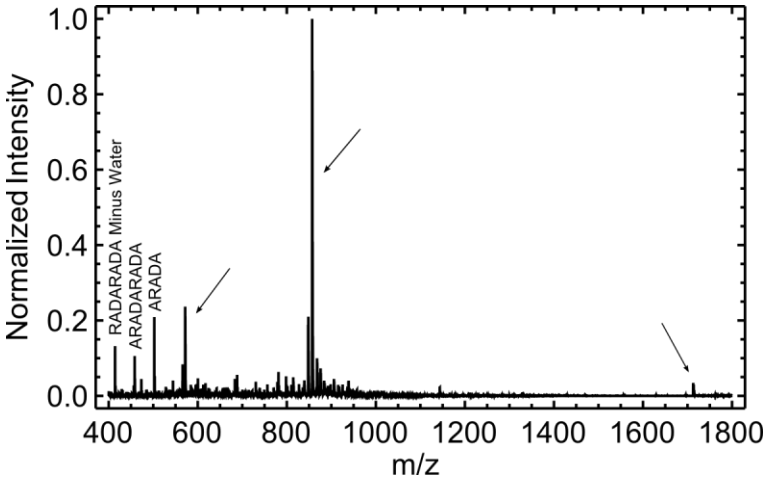

Figure S1: Electrospray ionization mass spectrum of RADA-F before HPLC purification. Arrows mark peaks which correspond to the desired RADA16-I peptide: m/z 429, 572, 857, and 1712. RADA16-I monoisotopic mass is 1712 Da. Other labeled peaks are assigned to C-terminal fragments.

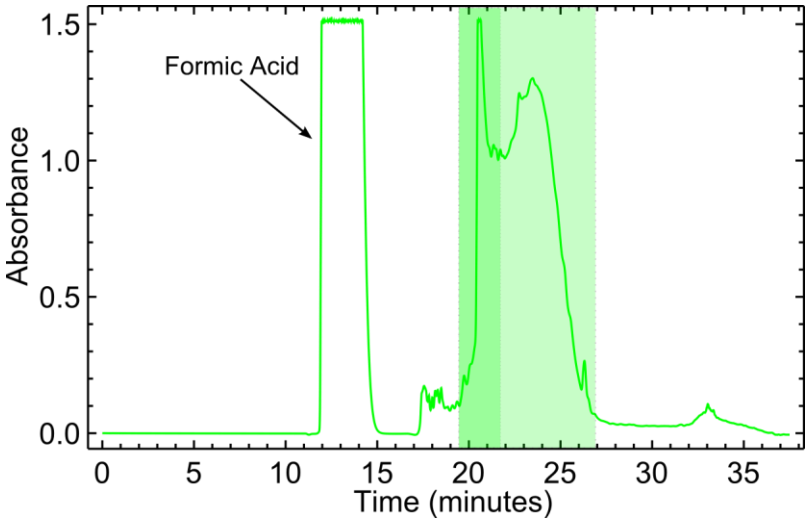

Figure S2: A sample HPLC trace for RADA-FA. The first main peak between 12-15 minutes is from formic acid, as verified by running with formic acid solution without peptide. The shaded areas indicate the collected fractions, which were combined later because they showed similar profiles by mass spectrometry. We suggest that the broad shape of this HPLC elution profile is due to self-association of the peptide.

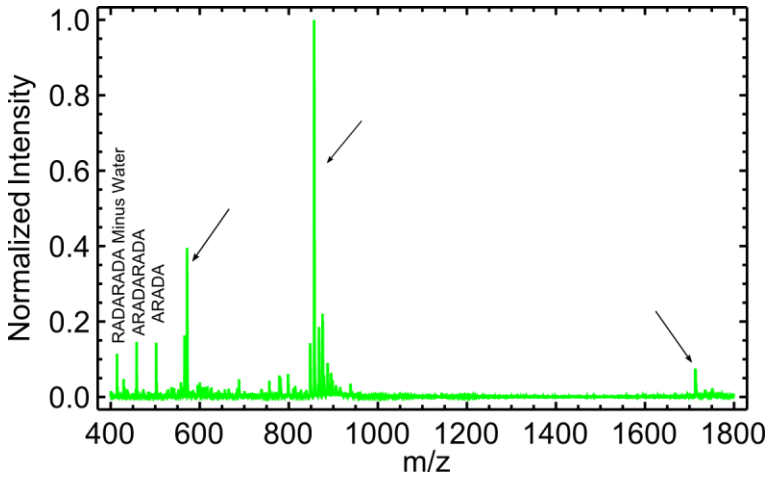

Figure S3: Electrospray ionization mass spectrum of RADA-FA sample after HPLC purification. Arrows mark peaks which correspond to the desired RADA16-I peptide: m/z 429, 572, 857, and 1712. RADA16-I monoisotopic mass is 1712 Da. Other labeled peaks are assigned to C-terminal fragments.

Consideration of Sample Heating During NMR Acquisition

We note that ssNMR measurements can heat samples due to absorption of radio frequency fields, primarily during ^{1}H decoupling, and friction during MAS. This concern was addressed by measuring RADA-F sample at a reduced MAS speed (12 kHz) and a lower ^{1}H decoupling power (50 kHz) at a 25 °C set point temperature. Under these conditions, we would expect substantially lower sample heating during NMR experiments. In Supplemental Figure 4a, we compare ssNMR spectra taken at 12 kHz MAS/50 kHz ^{1}H decoupling to spectra taken at 12 kHz MAS/110 kHz ^{1}H decoupling and 25 kHz MAS/110 kHz ^{1}H decoupling. These ssNMR spectra showed similar positions and lineshapes for carbonyl, α-, and alanine β-carbon signals, and no evidence indicating that unintentional heating during ssNMR measurements was large enough to induce structural changes in the sample. We also acquired a CPMAS ssNMR spectrum of RADA-F at a set point of -10 °C and 11 kHz MAS/110 kHz ^{1}H decoupling and compared this to a CPMAS NMR spectrum of the same sample at a set point of 25 °C and 11 kHz MAS/110 kHz ^{1}H decoupling (Supplemental Figure 4b).

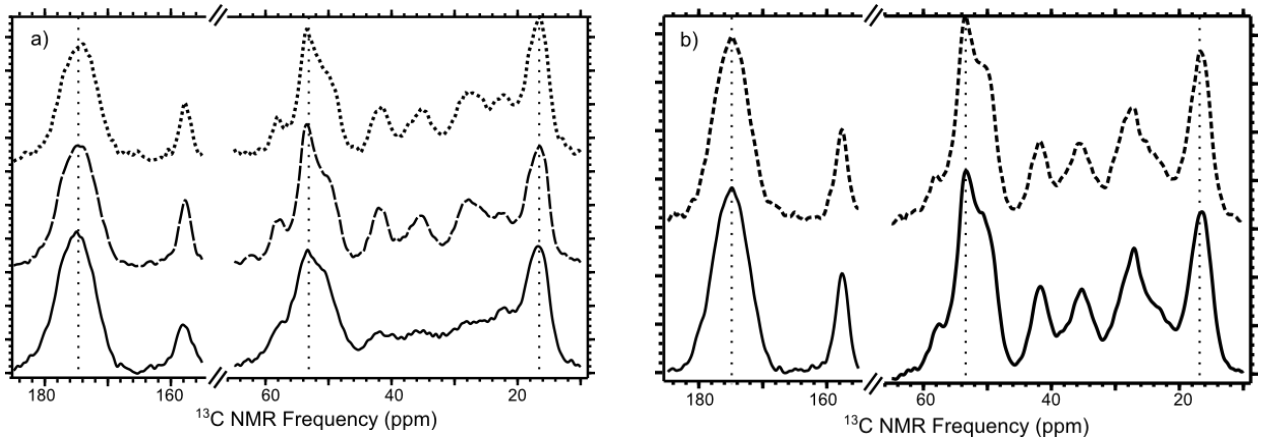

Figure S4: CPMAS ssNMR spectra of RADA-F sample acquired at variable MAS spinning speed, ^{1}H decoupling powers, and set point temperature, to rule-out effects on secondary structure of undesired heating during NMR acquisition. Vertical dotted lines are guides to the eye and indicate that we observed no change in peak positions for carbonyl, α-carbon, and alanine β-carbon signals. a) Spectra correspond to 50 kHz ^{1}H decoupling and 12 kHz MAS (solid), 110 kHz ^{1}H decoupling and 12 kHz MAS (dashed), and 110 kHz ^{1}H decoupling and 25 kHz MAS (dotted). b) CPMAS ssNMR spectra of a RADA-F sample acquired with 110 kHz ^{1}H decoupling and 11 kHz MAS, at -10 °C set point temperature (solid black) and 25 °C set point temperature (dashed black).

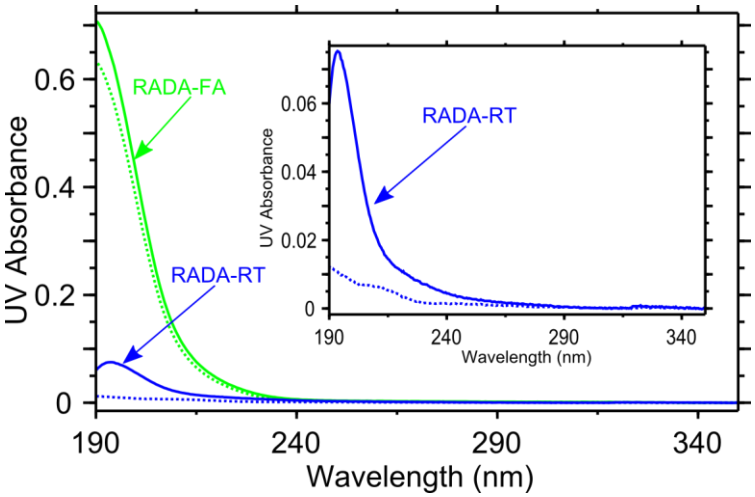

Figure S5: UV-Vis absorbances for RADA-FA and RADA-RT sonicated (solid) and not sonicated (dashed). The inset is an expansion of the RADA-RT data.

Table S1: Sample solubilities estimated by integrating UV-Vis spectra between 190 and 230 nm and scaled relative to the sonciated solubility reference sample (RADA-FA).

Sample	Percent Solubility Before Sonication	Percent Solubility After Sonication	Solubility Ratio, before vs. after sonication
RADA-F	8.9%	26%	2.9
RADA-RT	2.6%	13%	5.0
RADA-100C	3.7%	19%	5.1
RADA-150C	9.4%	18%	1.9
RADA-FA	88%	100%	1.1

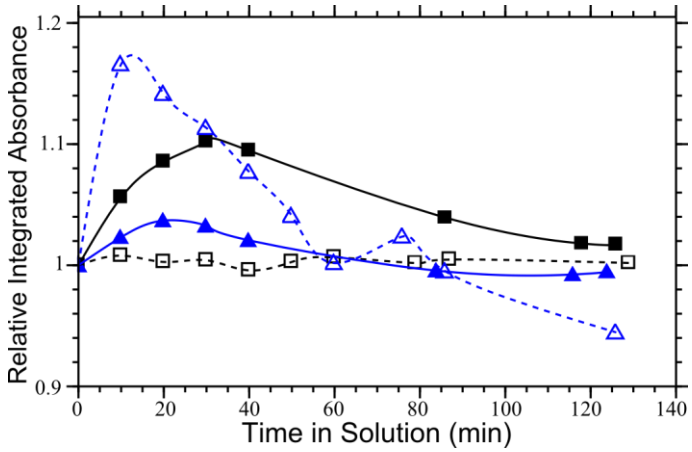

Figure S6: Time dependence of integrated UV absorbance between 190 and 230 nm, following sonication. Each data set is scaled to its initial value to show relative time dependence. Data sets correspond to RADA-F (black squares) and RADA-RT (blue triangles) solutions prepared with (solid lines, filled symbols) or without (dashed lines, unfilled symbols) sonication. The lines are guides to the eye.

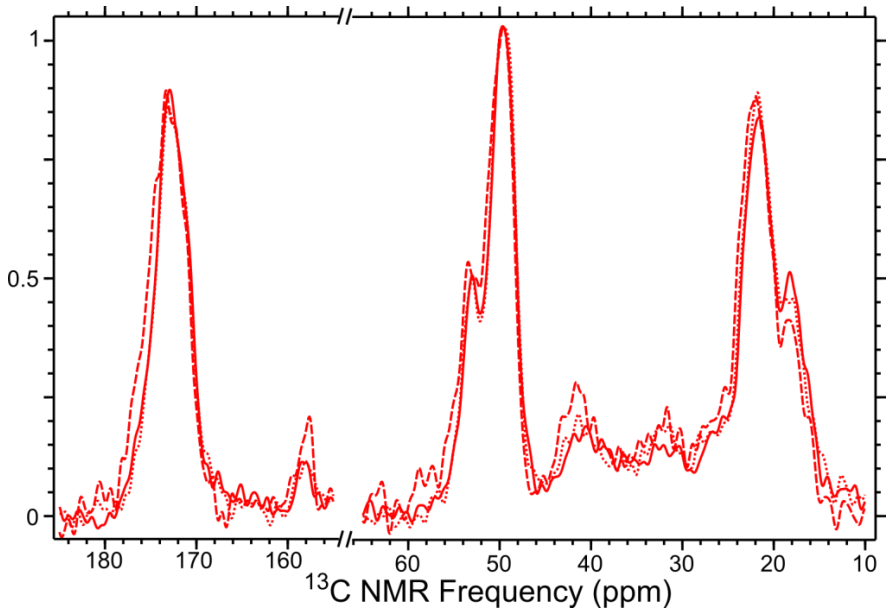

Figure S7: CPMAS ssNMR of RADA-80C immediately (solid) after being heated (12 hr acquire), the same sample measured the next morning after being allowed to cool (dotted), and the same sample 5 days later after being stored at room temperature (dashed).

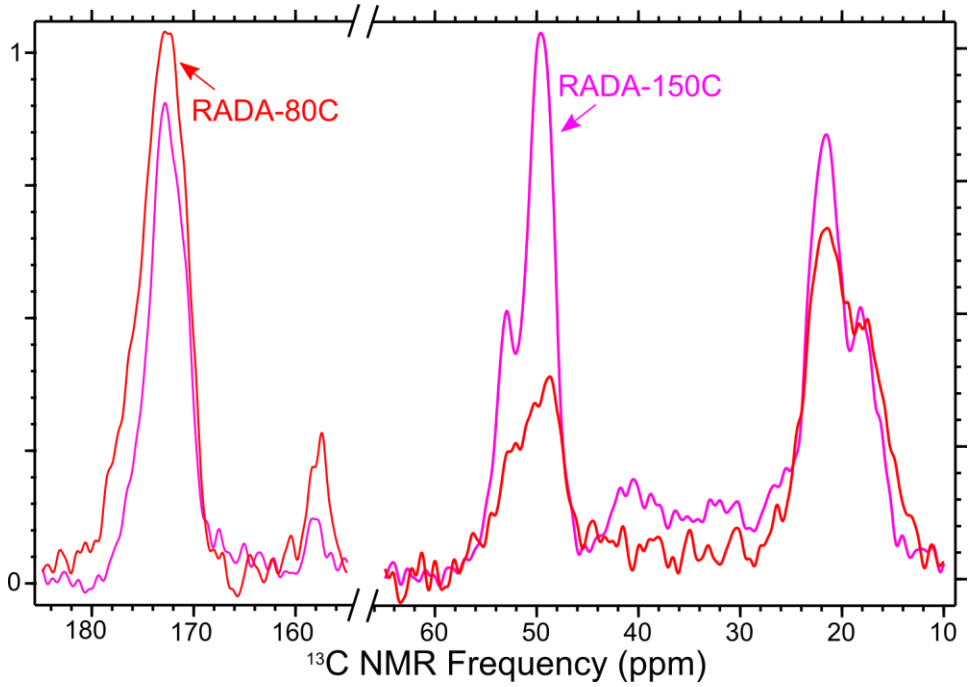

Figure S8: CPMAS ssNMR of RADA-80C (red) heated by the ssNMR spectrometer and RADA-150C (magenta) heated in DSC.