

## Supplementary Information

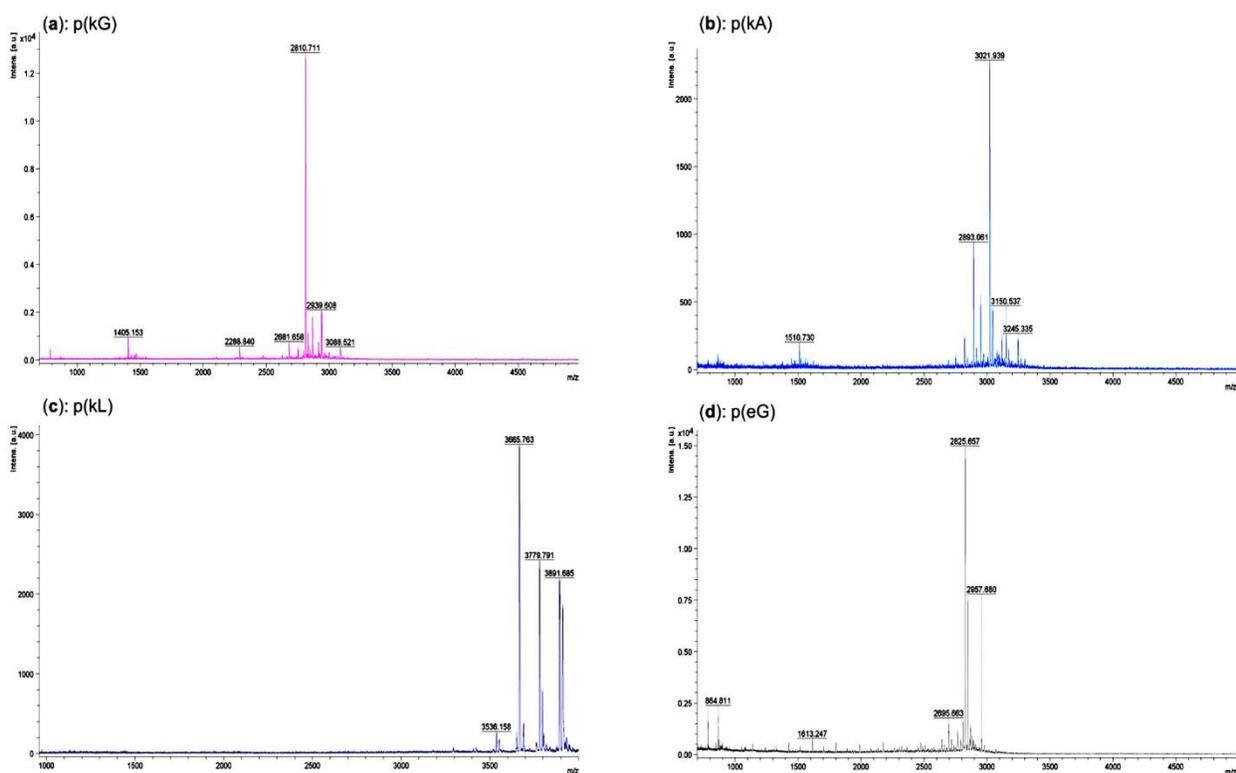
# Engineering Peptide-based Polyelectrolyte Complexes with Increased Hydrophobicity

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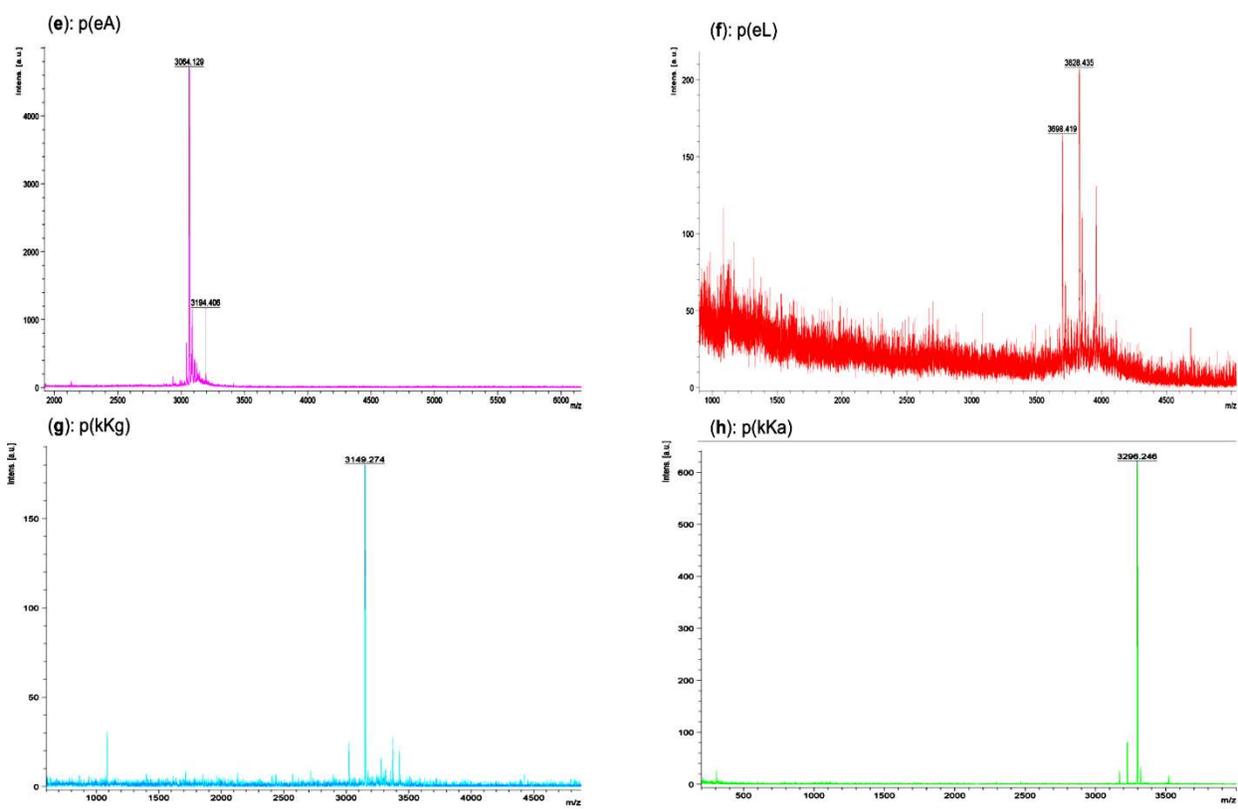
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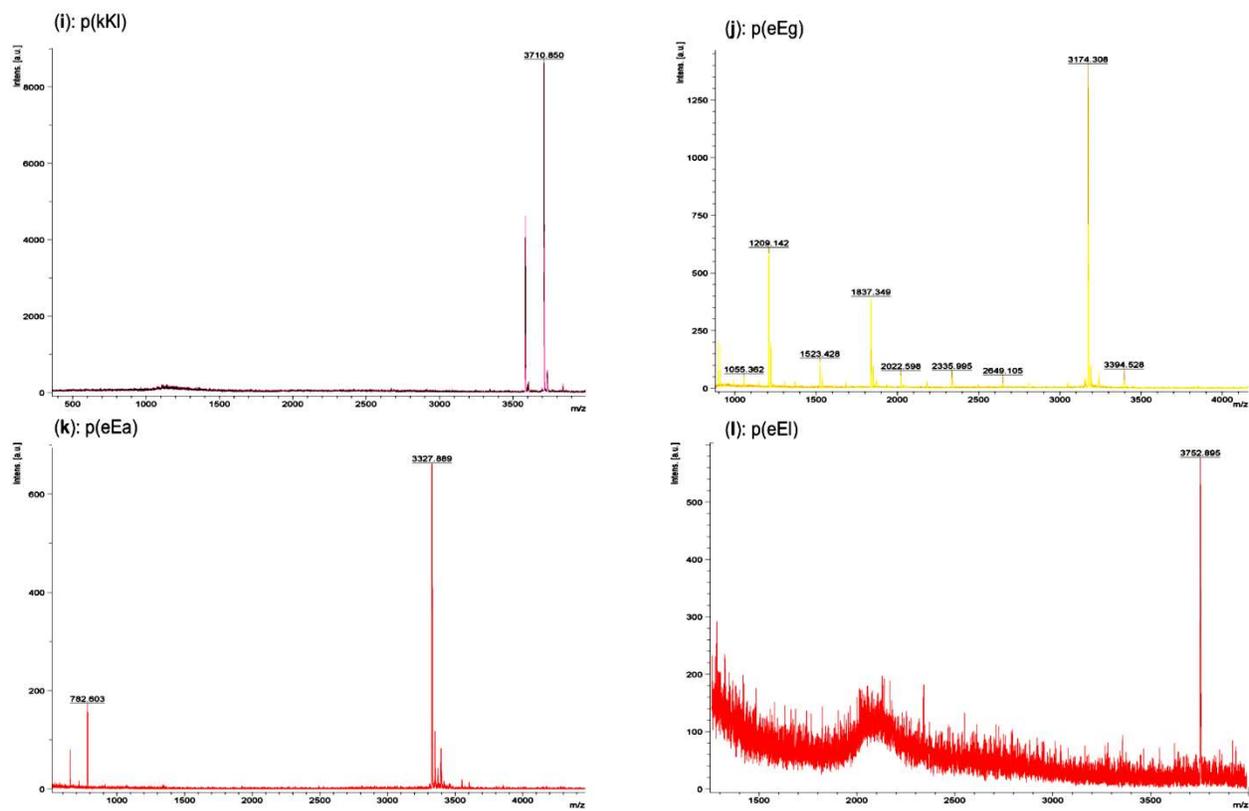
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**Figure S1.** MALDI-TOF mass spectroscopy of the peptide sequences: (a) p(kG); (b) p(kA); (c) p(kL); (d) p(eG).

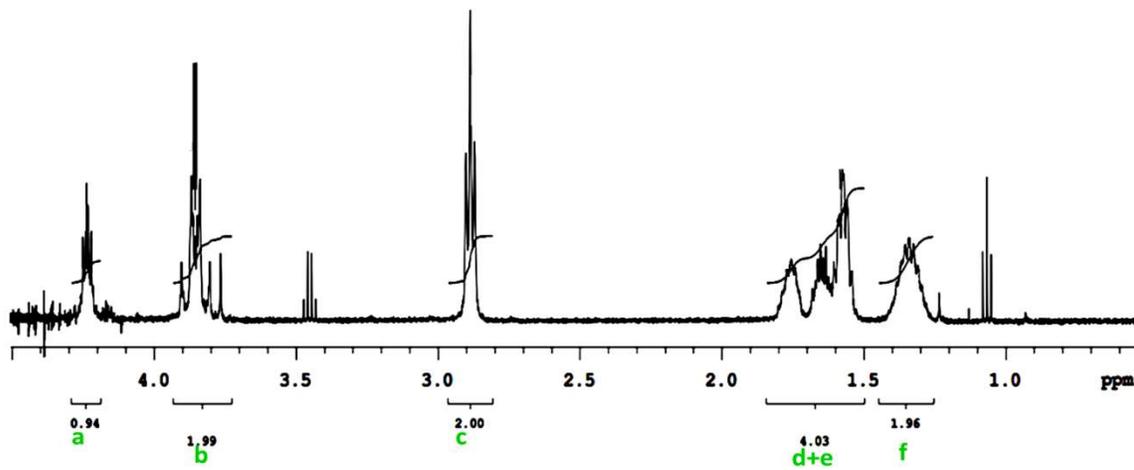
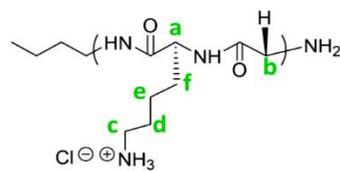


**Figure S1. Continued.** MALDI-TOF mass spectroscopy of the peptide sequences: (e) p(eA); (f) p(eL); (g) p(kKg); (h) p(kKa).



**Figure S1. Continued.** MALDI-TOF mass spectroscopy of the peptide sequences: (i) p(kKl); (j) p(eEg); (k) p(eEa); (l) p(eEl).

p(kG)



p(eG)

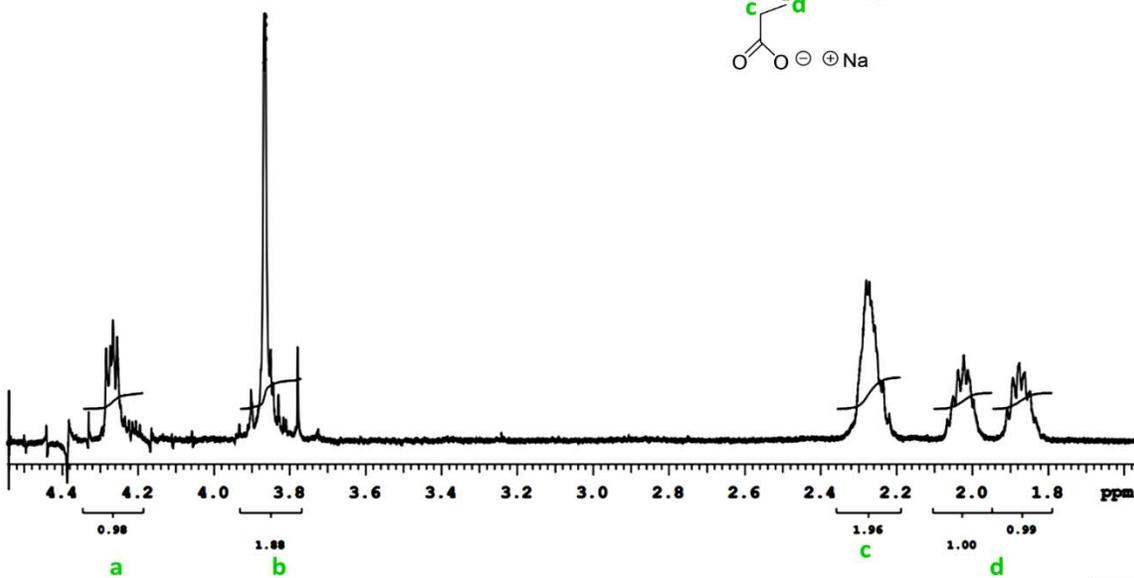
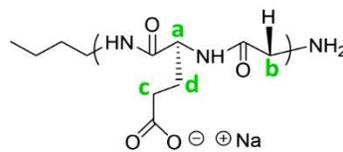
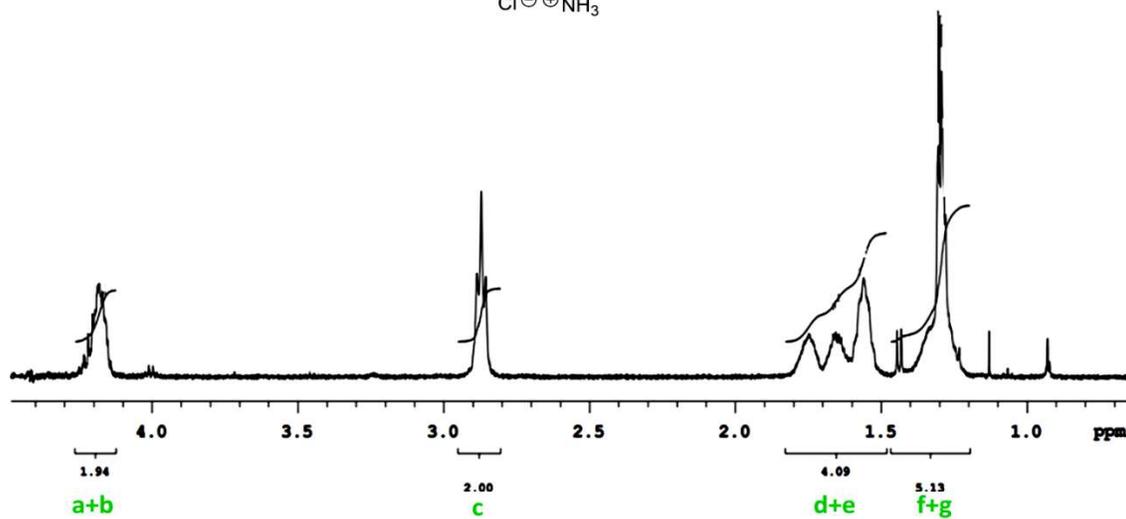
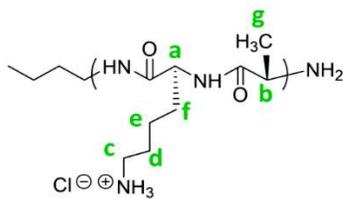


Figure S2.  $^1\text{H}$  NMR spectroscopy of the peptide sequences: p(kG) on top; p(eG) on bottom.

p(kA)



p(eA)

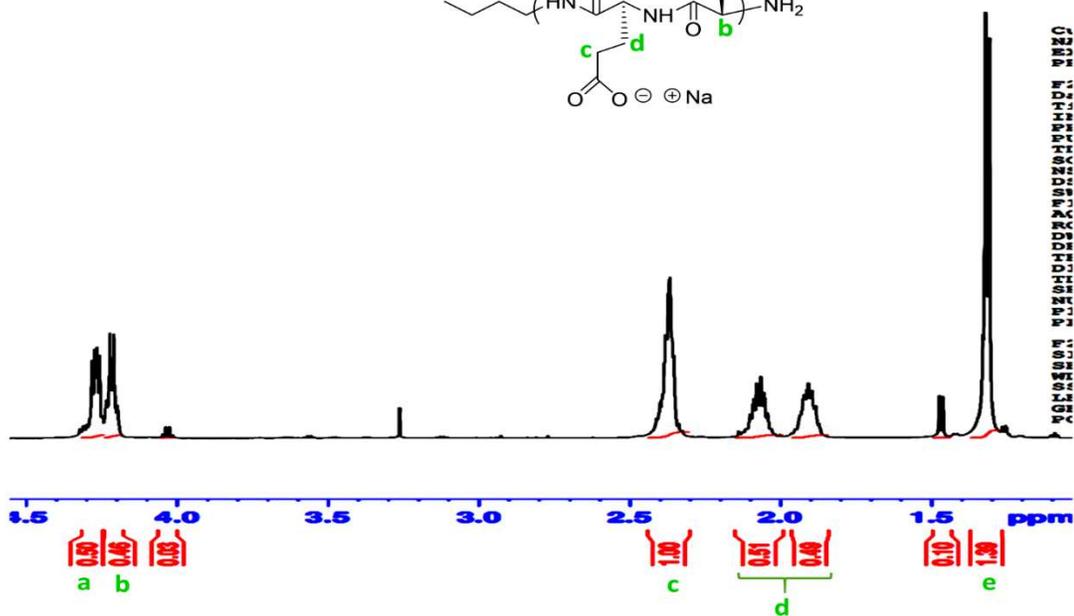
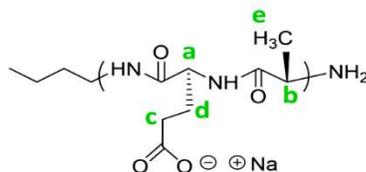
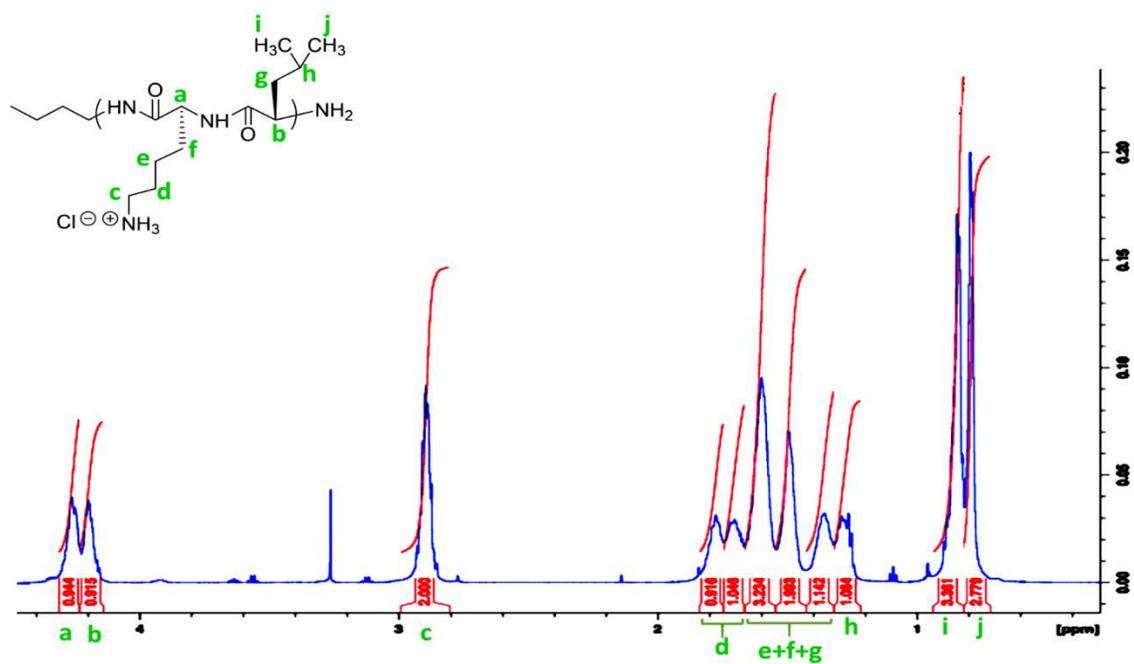


Figure S2. Continued.  $^1\text{H}$  NMR spectroscopy of the peptide sequences: p(kA) on top; p(eA) on bottom.

p(kL)



p(eL)

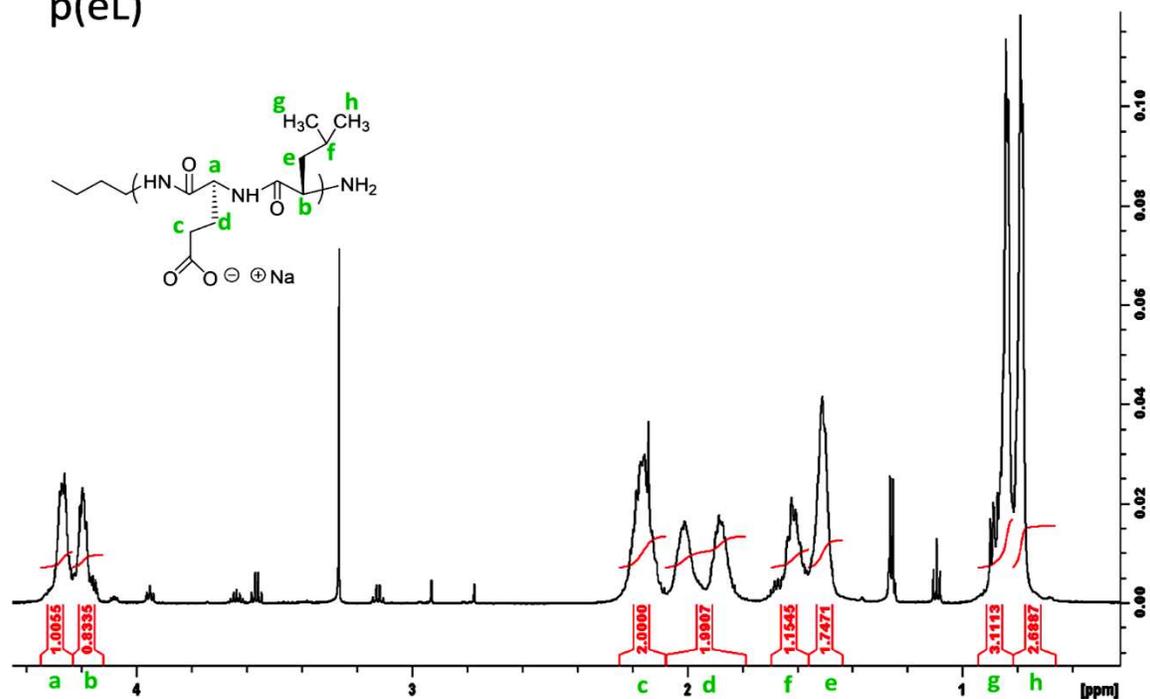
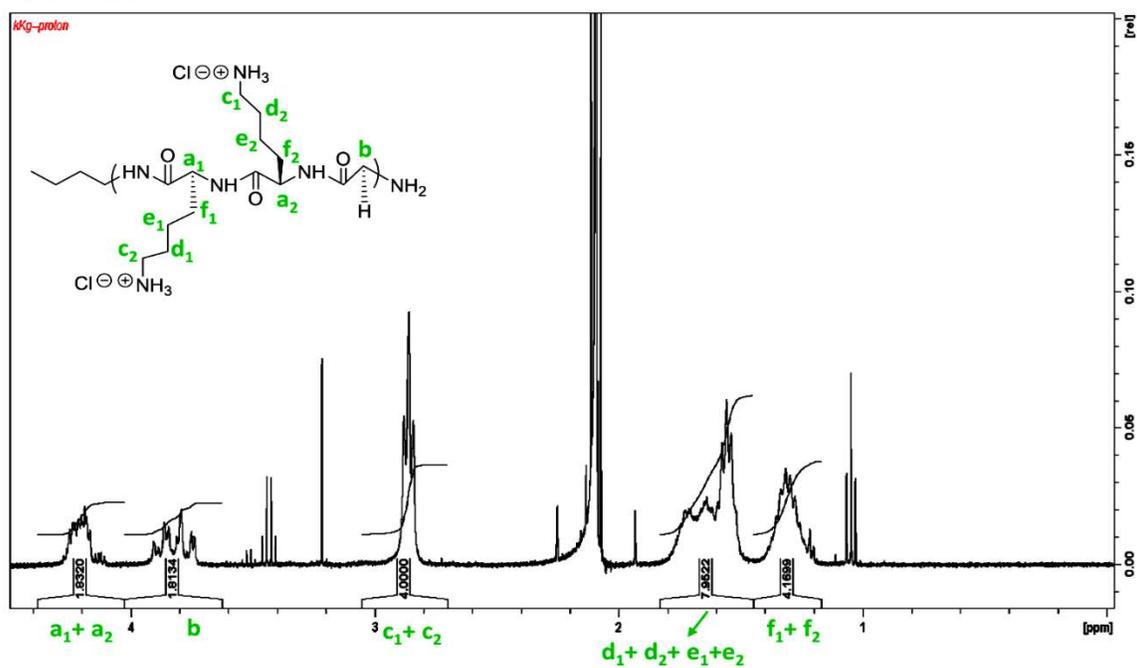


Figure S2. Continued. <sup>1</sup>H NMR spectroscopy of the peptide sequences: p(kL) on top; p(eL) on bottom.

p(kKg)



p(eEg)

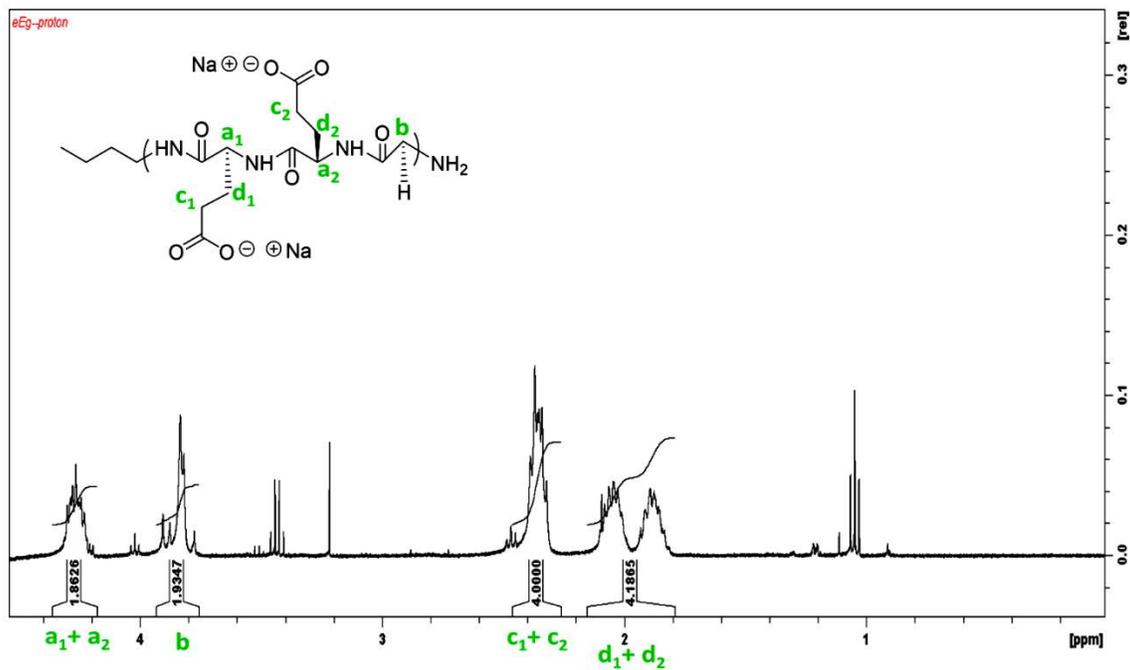
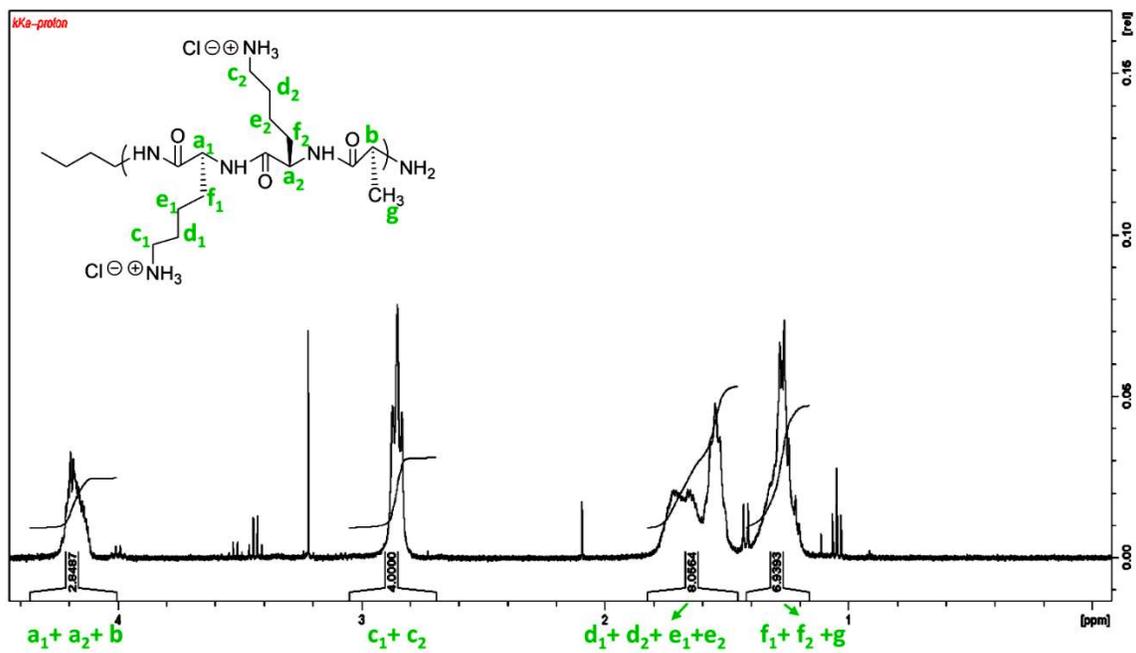


Figure S2. Continued.  $^1H$  NMR spectroscopy of the peptide sequences: p(kKg) on top; p(eEg) on bottom.

## p(kKa)



## p(eEa)

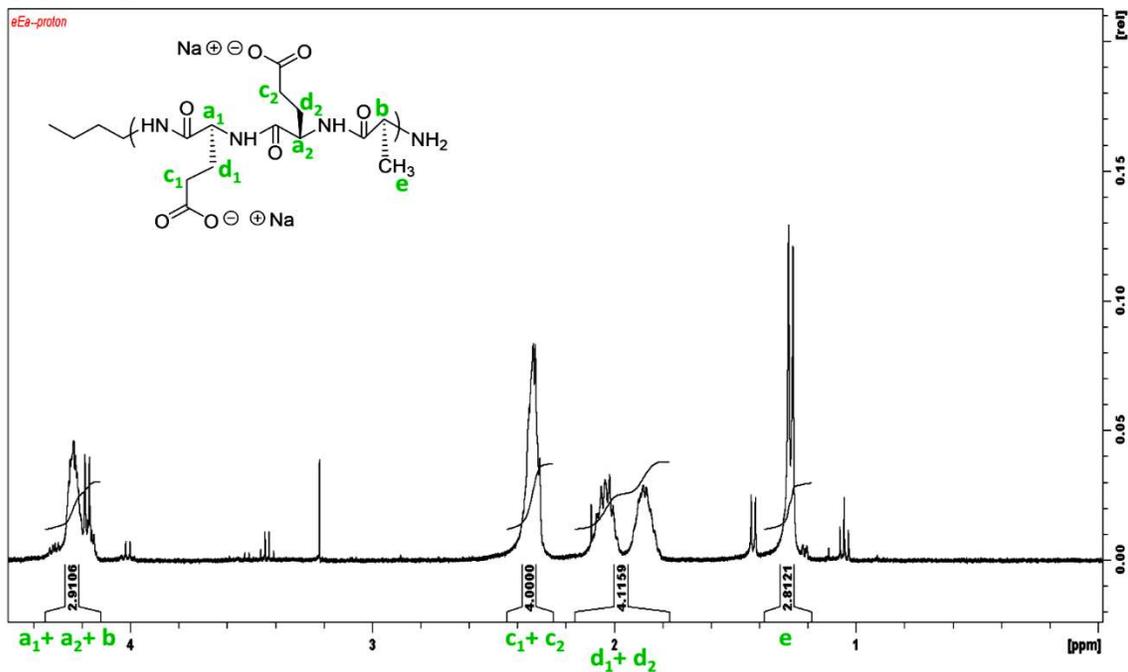
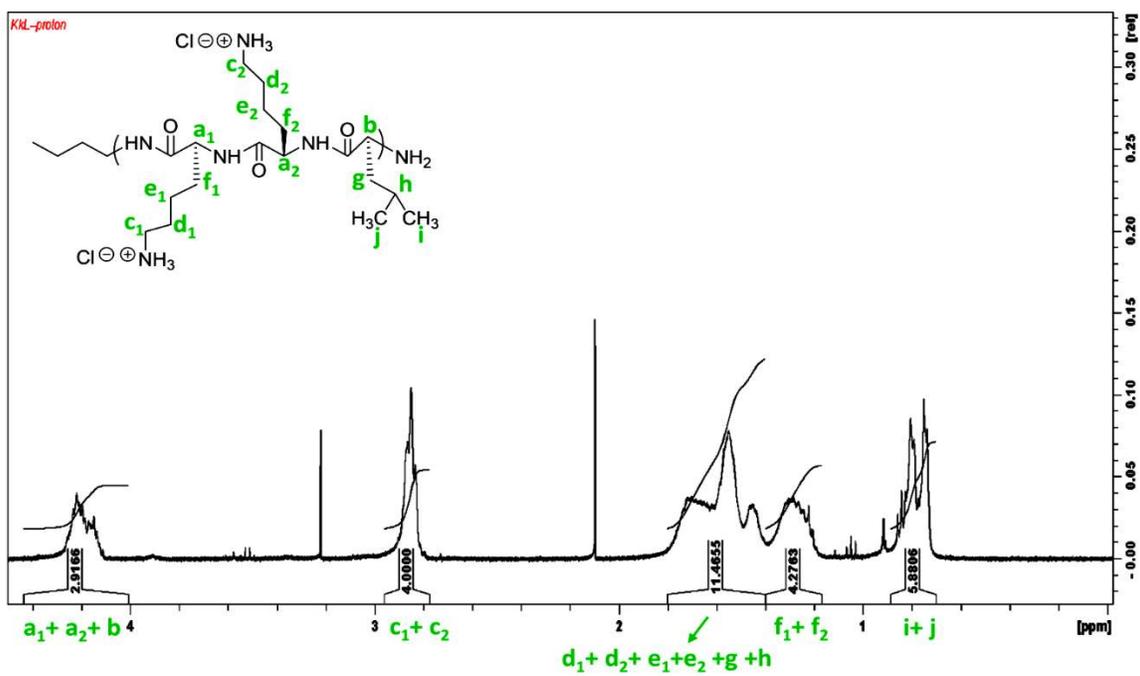


Figure S2. Continued.  $^1H$  NMR spectroscopy of the peptide sequences: p(kKa) on top; p(eEa) on bottom.

### p(kKI)



### p(eEI)

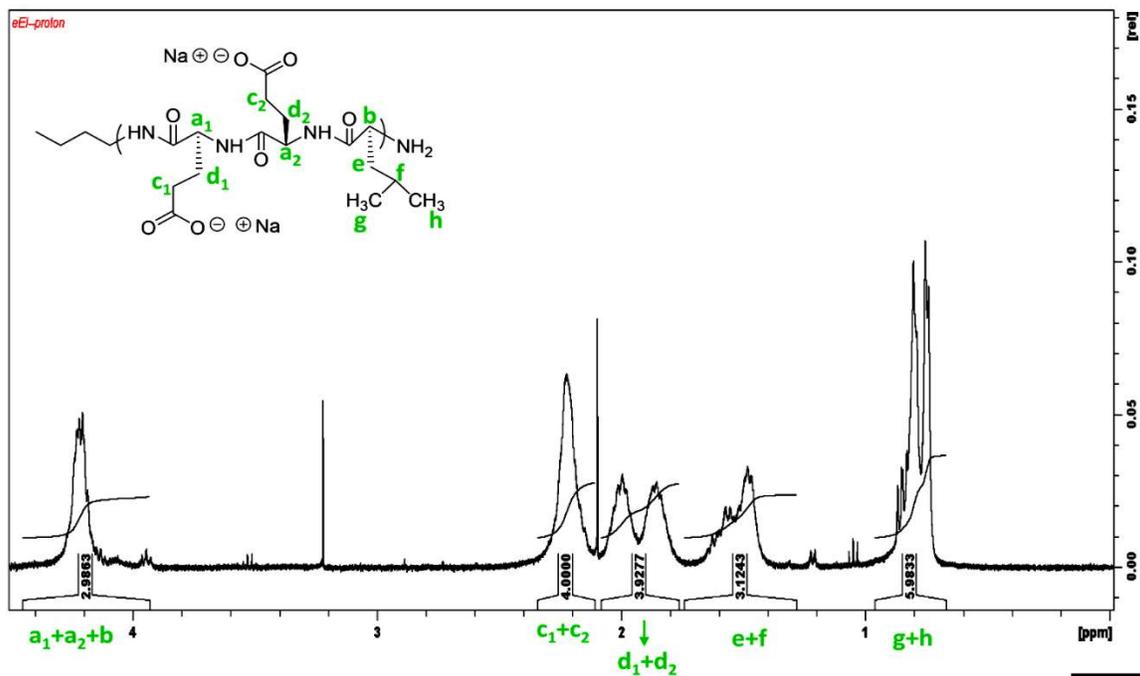
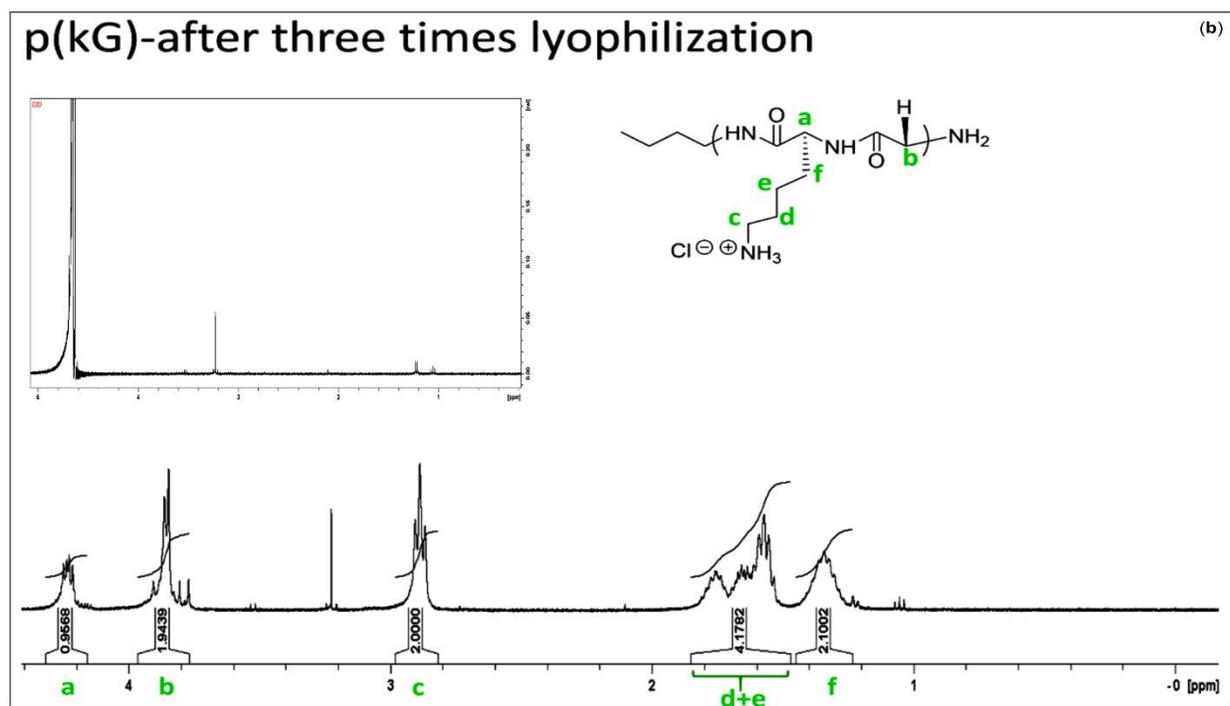
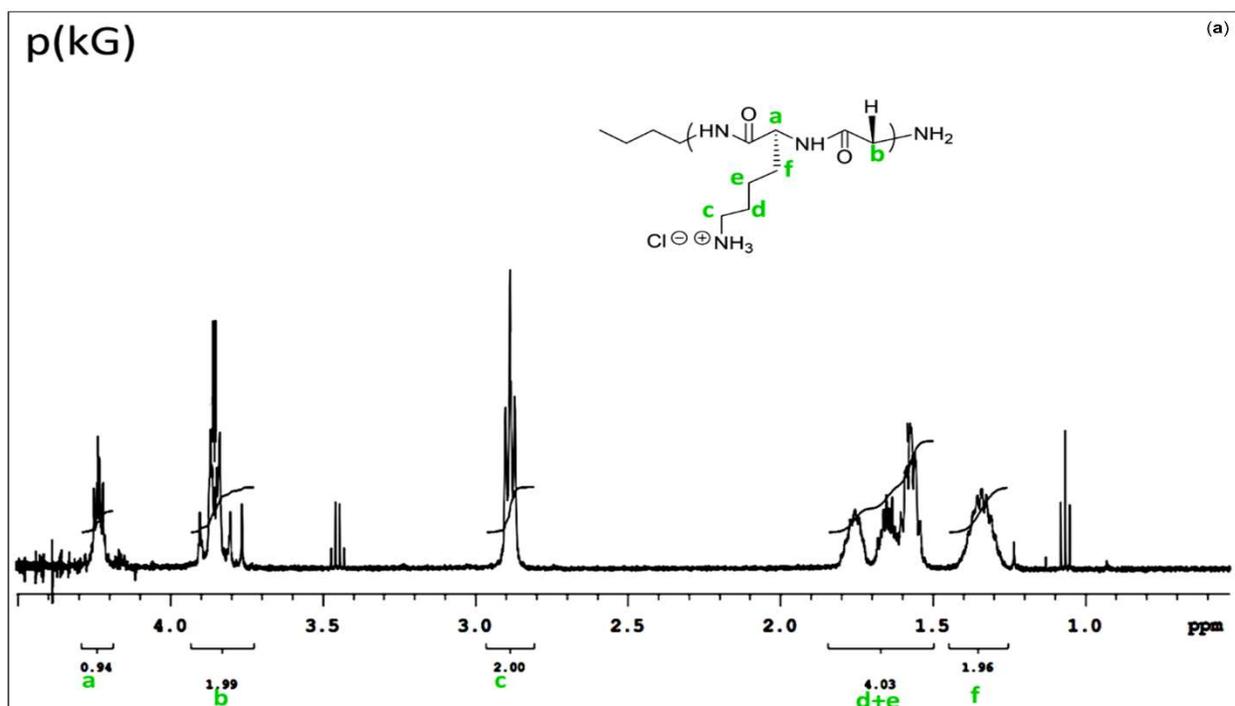
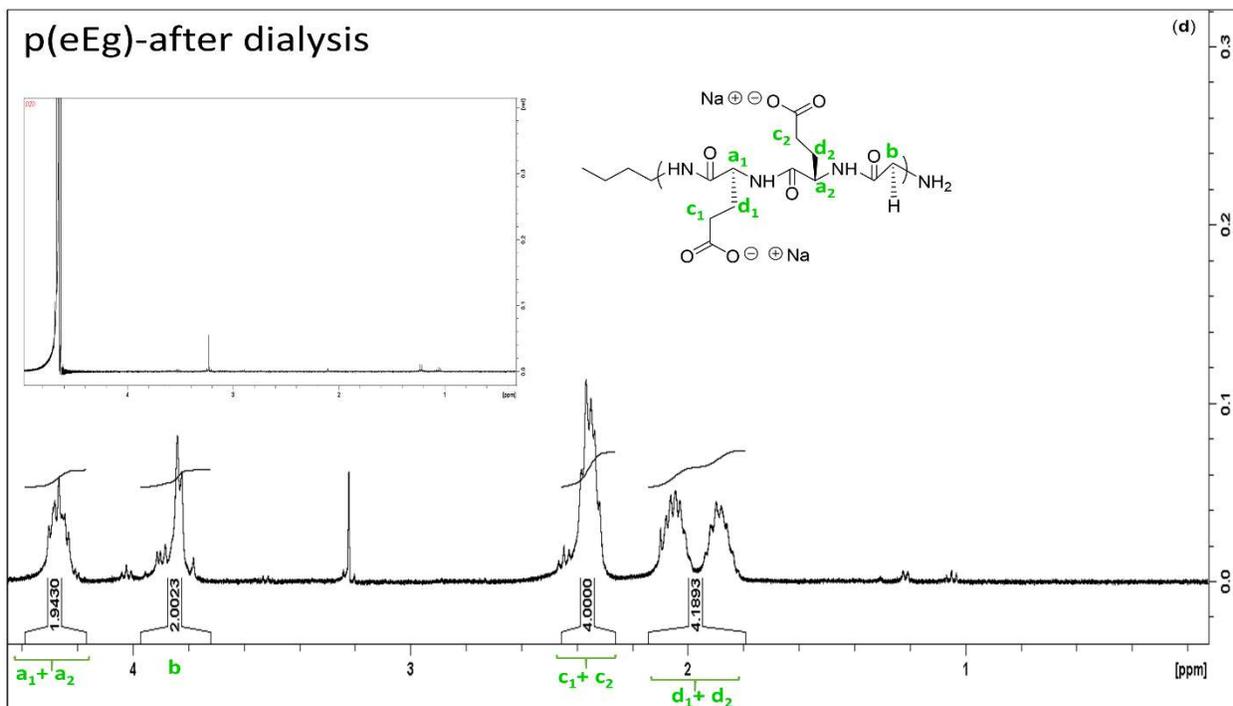
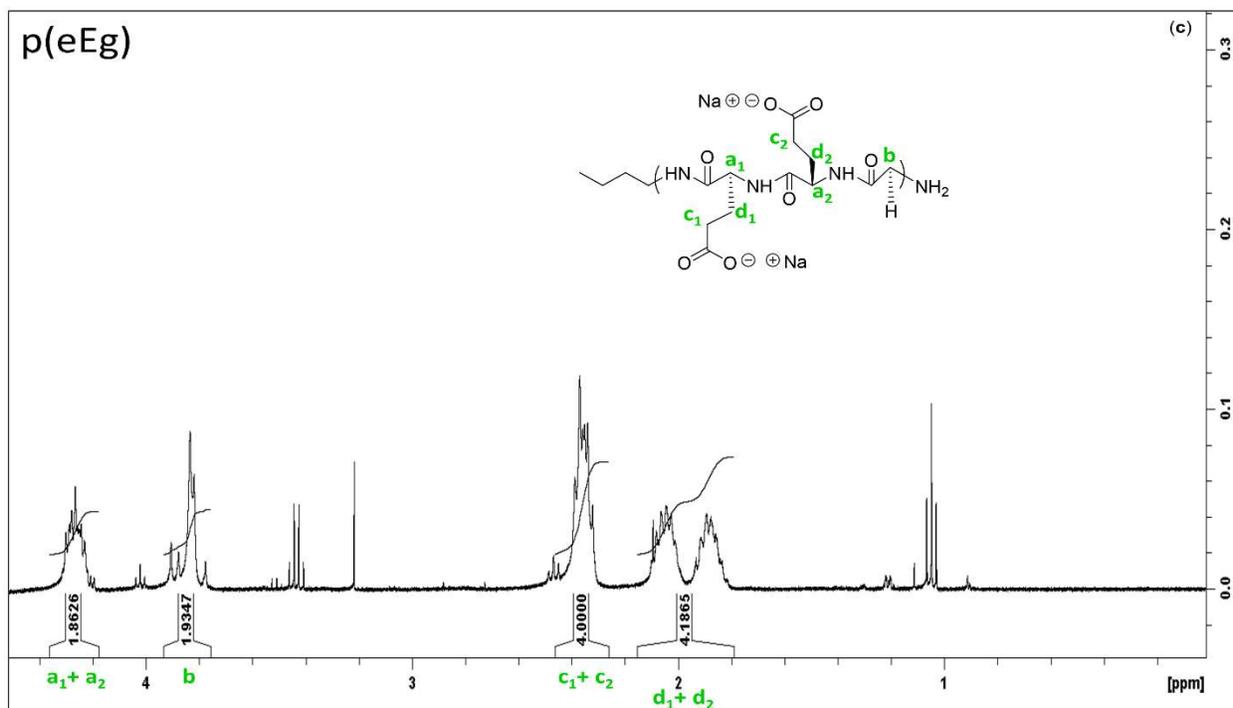


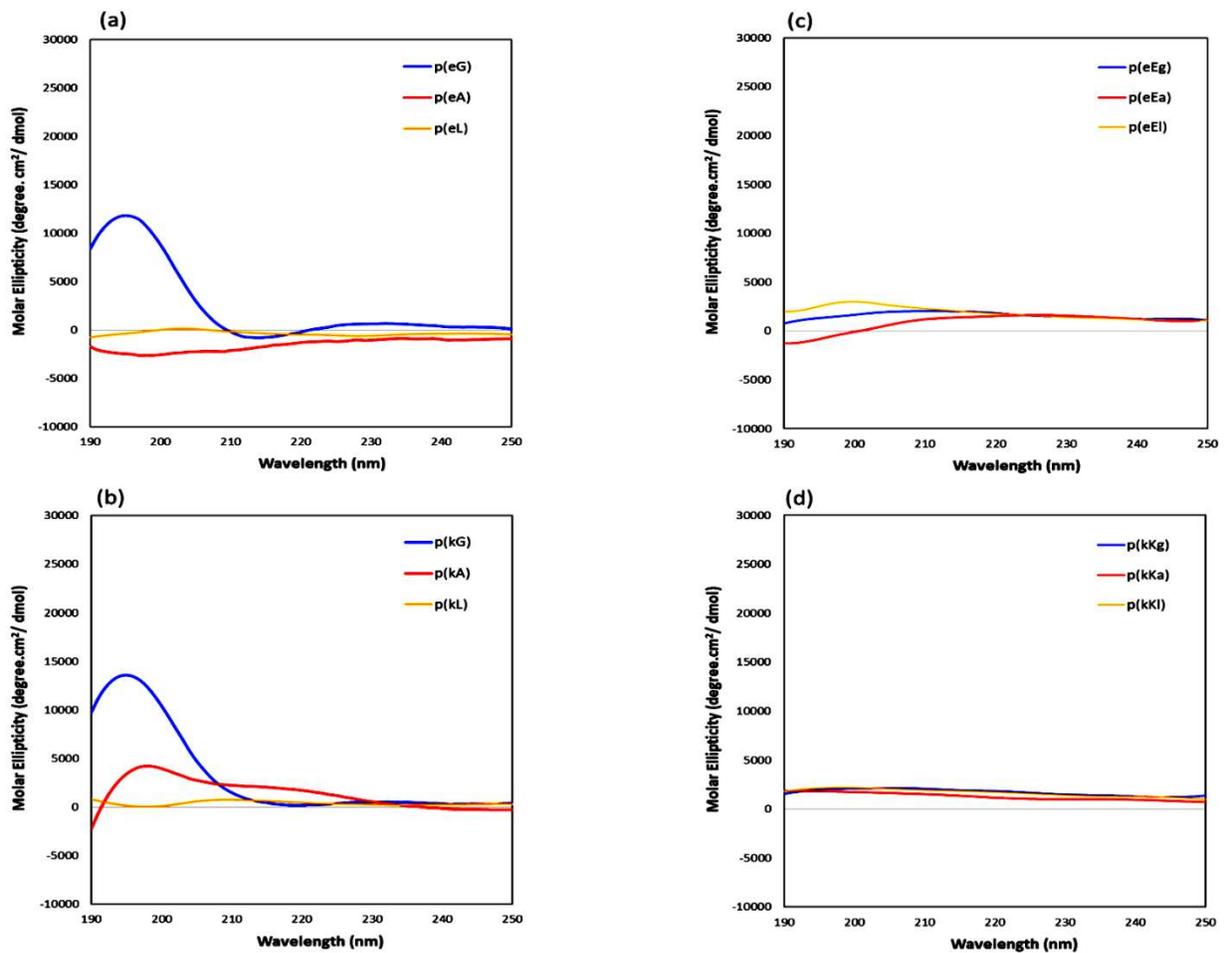
Figure S2. Continued.  $^1H$  NMR spectroscopy of the peptide sequences: p(kKI) on top; p(eEI) on bottom.



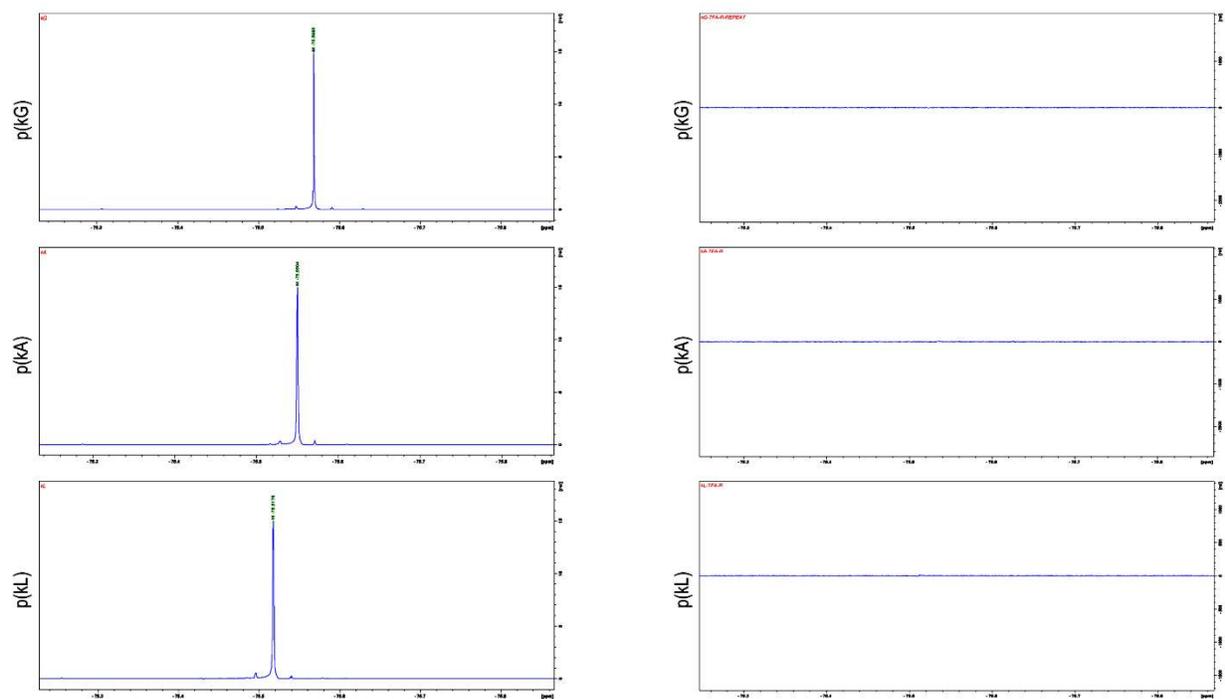
**Figure S3.**  $^1\text{H}$  NMR spectroscopy of polypeptides: (a) p(kG) which shows the signals of diethyl ether at 1.17 and 3.56 ppm for  $\text{CH}_3$  and  $\text{CH}_2$ , respectively; (b) p(kG) after three times lyophilization indicating removal of diethyl ether. Inset:  $^1\text{H}$  NMR spectrum of  $\text{D}_2\text{O}$ . The signals around 1 and 3.2 ppm can also be observed in the spectrum of  $\text{D}_2\text{O}$  alone which was used as the solvent for the NMR measurements.



**Figure S3. Continued.**  $^1\text{H}$  NMR spectroscopy of polypeptides: (c) p(eEg) which shows the signals of diethyl ether at 1.17 and 3.56 ppm for  $\text{CH}_3$  and  $\text{CH}_2$ , respectively; (d) p(eEg) after dialysis indicating removal of diethyl ether. Inset:  $^1\text{H}$  NMR spectrum of  $\text{D}_2\text{O}$ . The signals around 1 and 3.2 ppm can also be observed in the spectrum of  $\text{D}_2\text{O}$  alone which was used as the solvent for the NMR measurements.



**Figure S4.** Circular dichroism (CD) spectroscopy of the peptide sequences: (a) p(eX); (b) p(kX); (c) p(eEx); (d) p(kKx).



**Figure S5.** F NMR spectroscopy of the polycations of p(kX) sequence: left side of the image shows the spectra before TFA-elimination; right side of the image shows the spectra after TFA-elimination.