Electronic Supplementary Information

Protein Characterization. As reported, all peptide samples were purchased from GenScript Inc. (Piscataway, NJ). Peptides were synthesized by solid-phase synthesis, purified by reverse-phase HPLC, and verified by LCMS. Figures 1-6 are shown to verify sample identity and purity for AKA_2, Trp-cage, and Trpzip4.

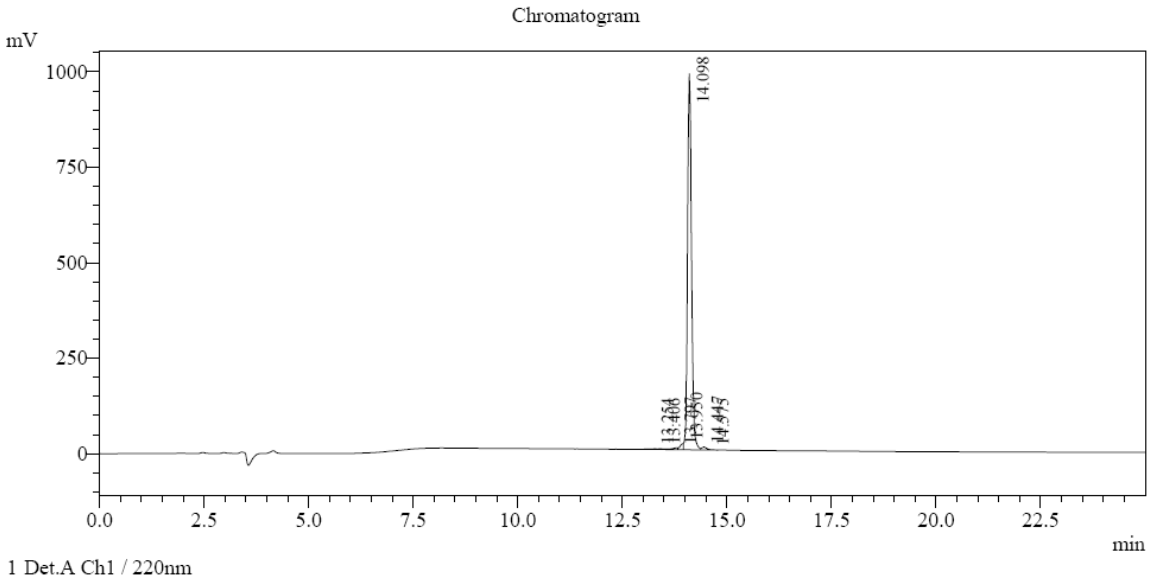

Figure 1. Reverse-phase HPLC Chromatogram verifying sample purity for AKA_2. Data provided by GenScript Inc.

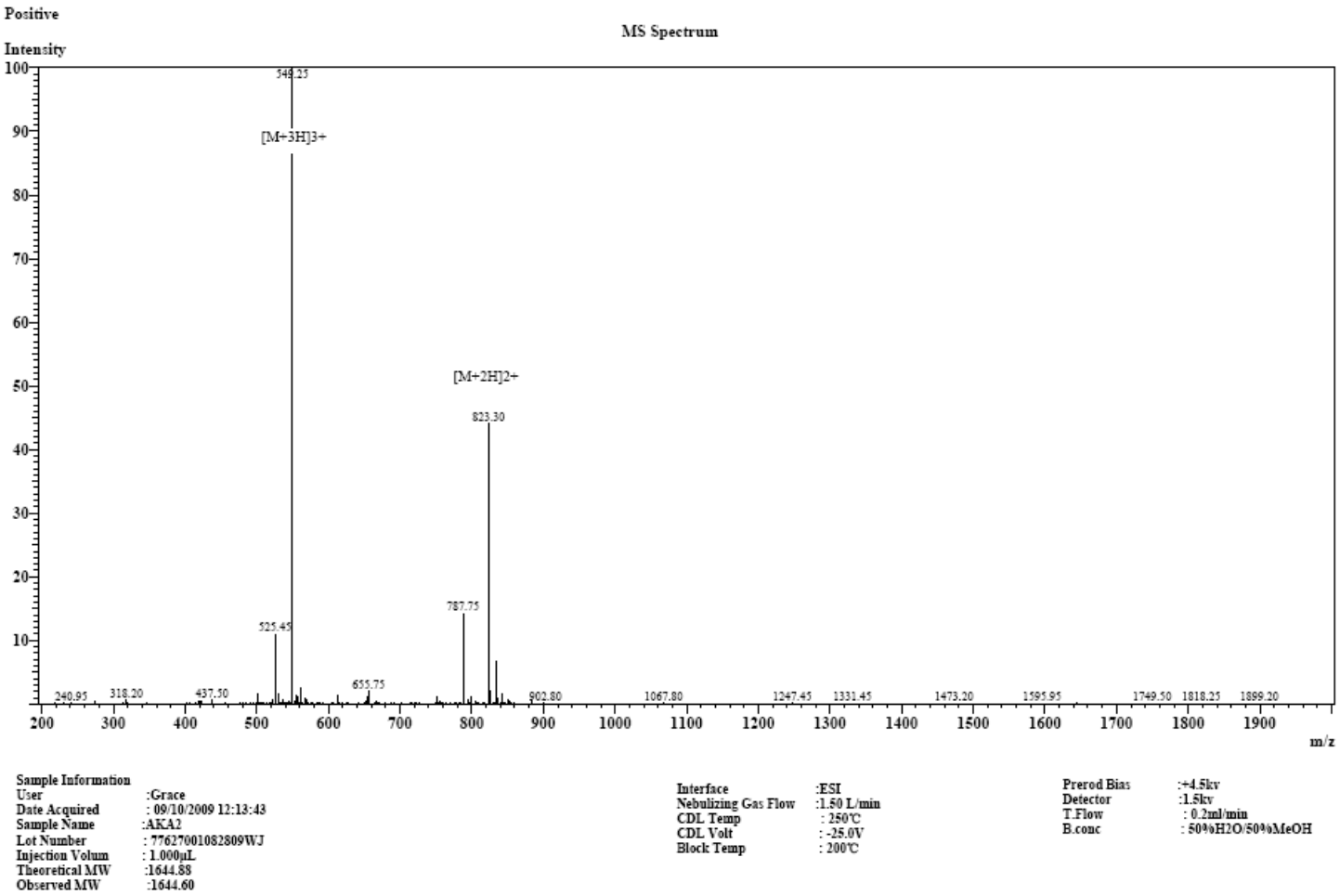

Figure 2. LCMS spectrum verifying sample identity for AKA_2. Data provided by GenScript Inc.

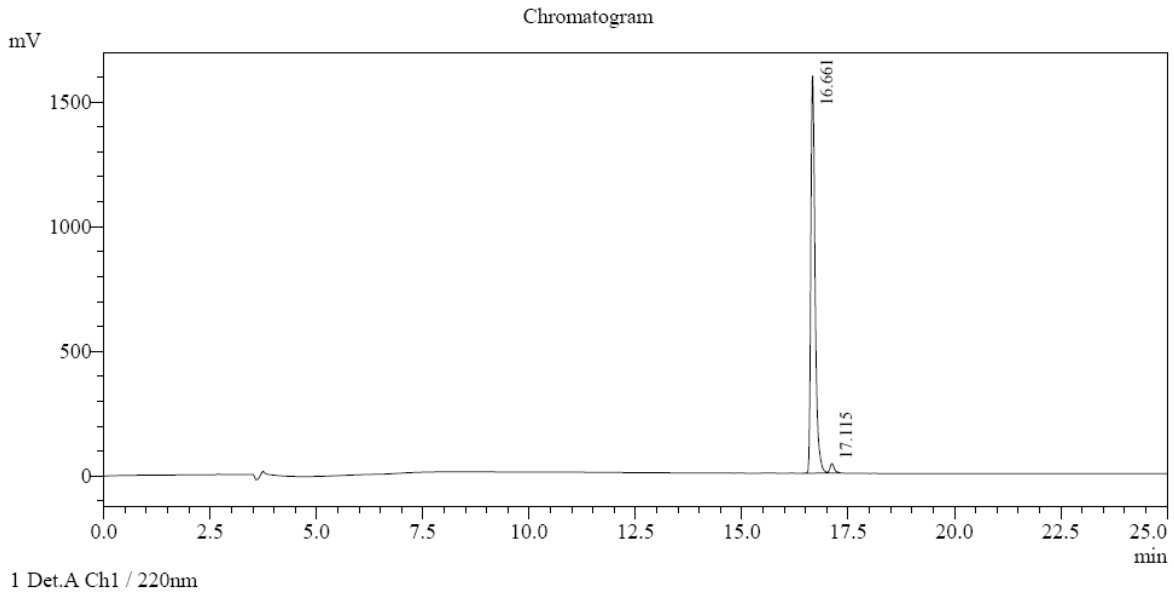

Figure 3. Reverse-phase HPLC Chromatogram verifying sample purity for Trp-cage. Data provided by GenScript Inc.

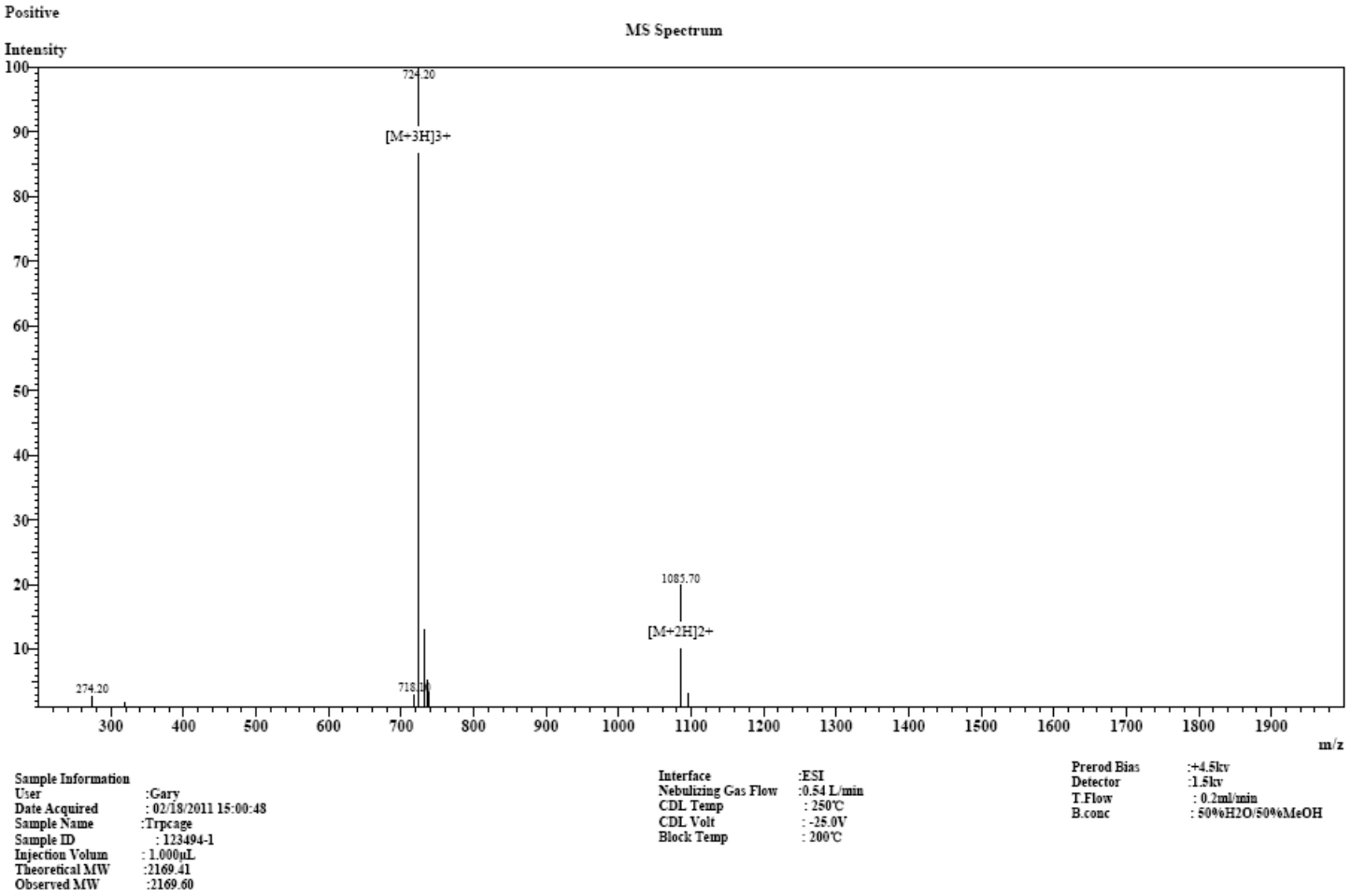

Figure 4. LCMS spectrum verifying sample identity for Trp-cage. Data provided by GenScript Inc.

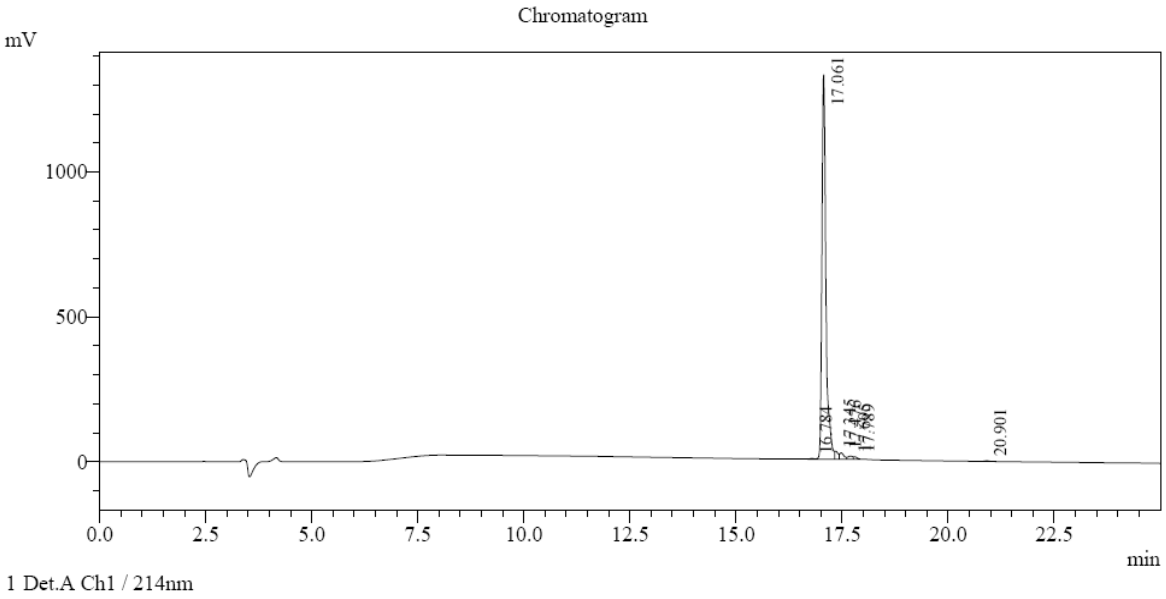

Figure 5. Reverse-phase HPLC Chromatogram verifying sample purity for Trpzip4. Data provided by GenScript Inc.

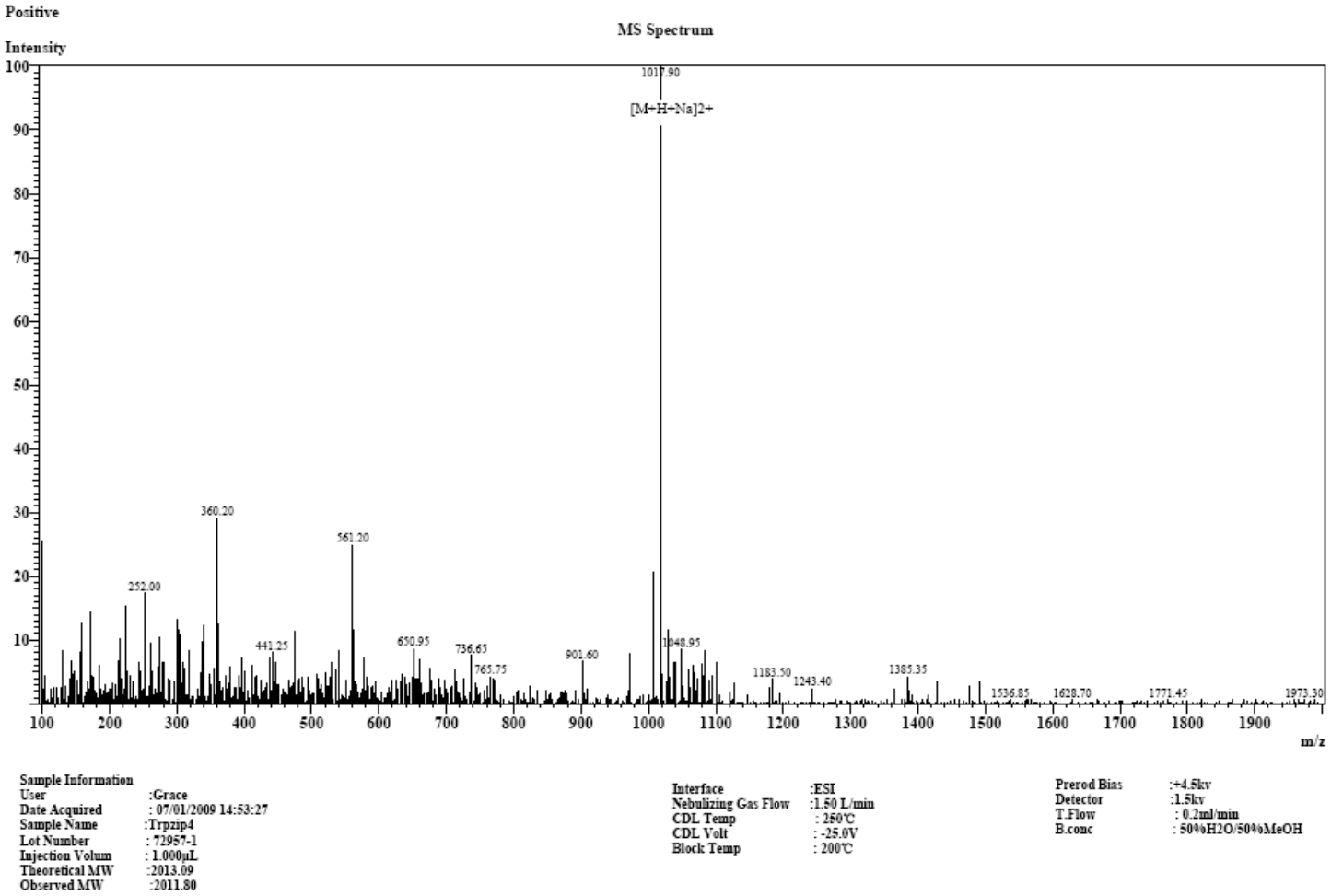

Figure 6. LCMS spectrum verifying sample identity for Trpzip4. Data provided by GenScript Inc.

UV Absorbance Data.

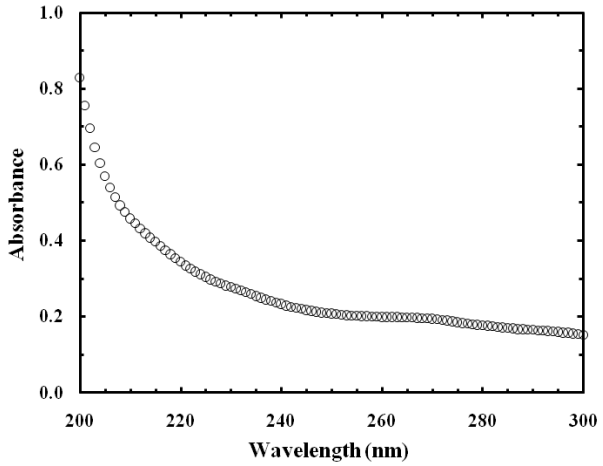

Figure 7. UV absorption spectrum of $[C_4mpy][Tf_2N]$, collected at room temperature.

Circular Dichroism Data.

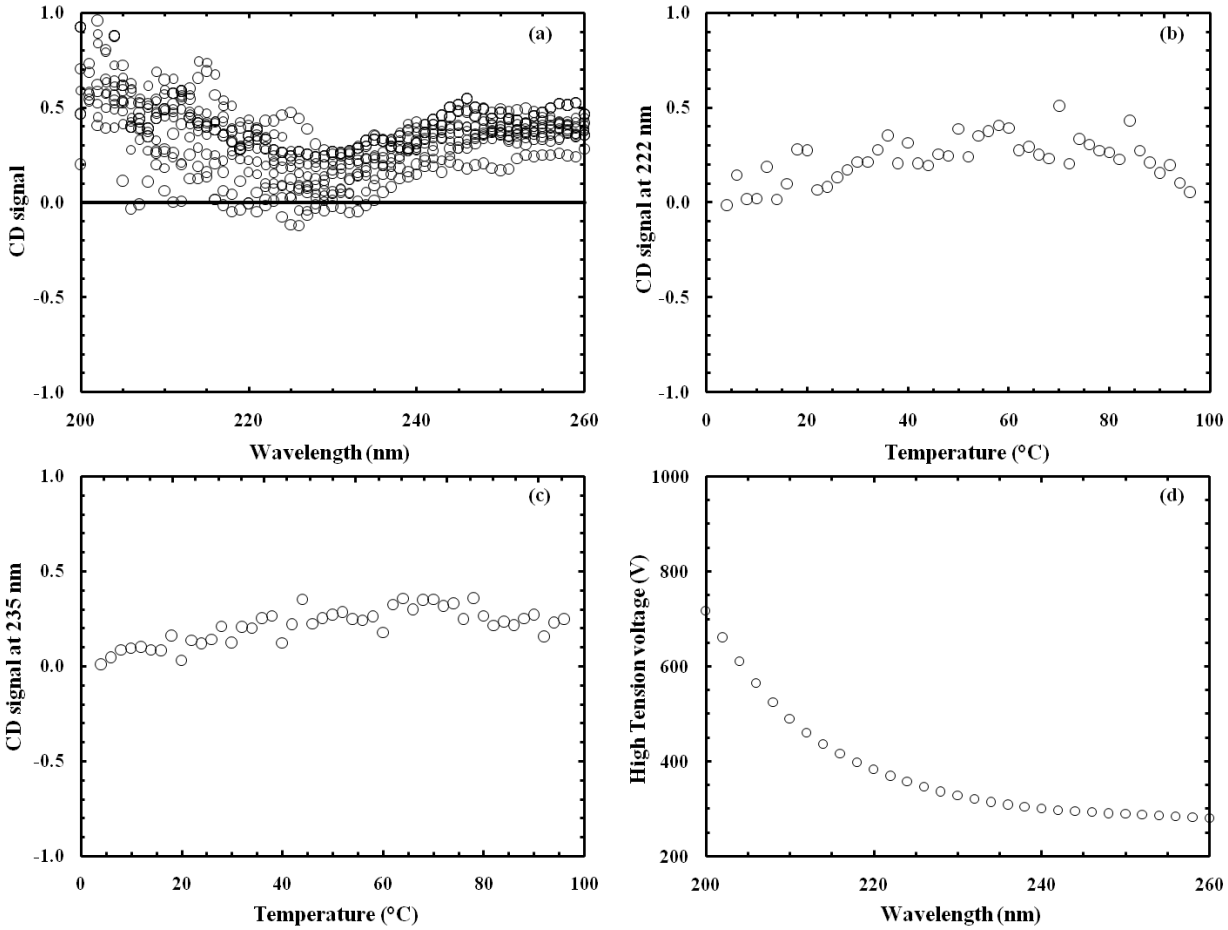

Figure 8. (a) $[C_4mpy][Tf_2N]$ far-UV spectra and CD signal at (b) 222 nm and (c) 235 nm as a function of temperature. The spectra in (a) are shown for 10 °C intervals from 4 °C to 94 °C. In general, the CD signal at a particular wavelength shows a slight increase with temperature. (d) Representative plot of CD high tension (HT) voltage for peptide samples in IL. Data shown corresponds to 100 μM Trpzip4 in $[C_4mpy][Tf_2N]$ and varies little with temperature. Literature suggests that reliable CD data can be obtained when the voltage is less than 700 V.[1] Temperature-induced changes in CD signal are monitored at 222 nm (AKA_2 or Trp-cage) or 235 nm (Trpzip4), where the HT voltage is less than 400 V.

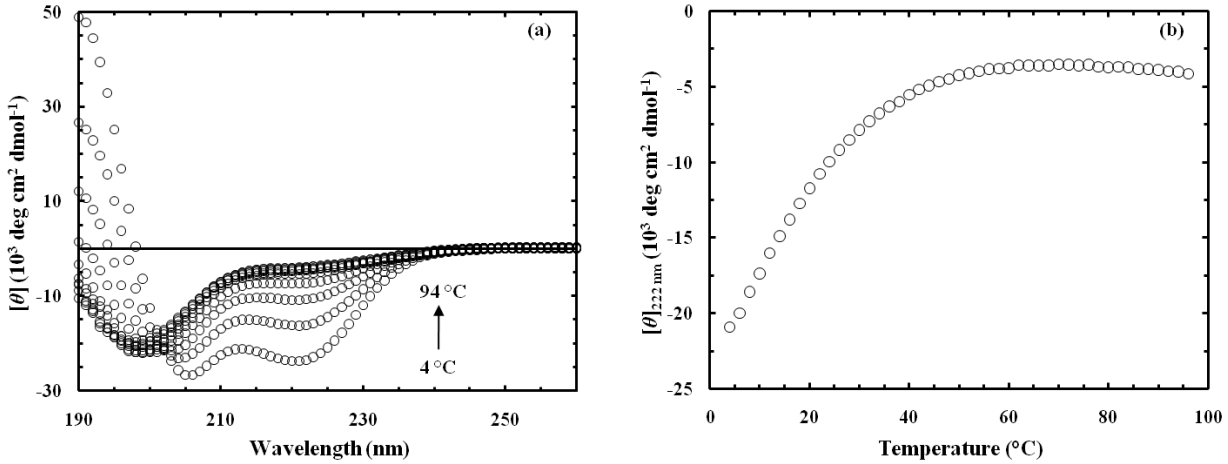

Figure 9. (a) Far-UV spectra and (b) mean residue ellipticity at 222 nm of AKA_2 in water as a function of temperature. The spectra in (a) are shown for 10 °C intervals from 4 °C to 94 °C.

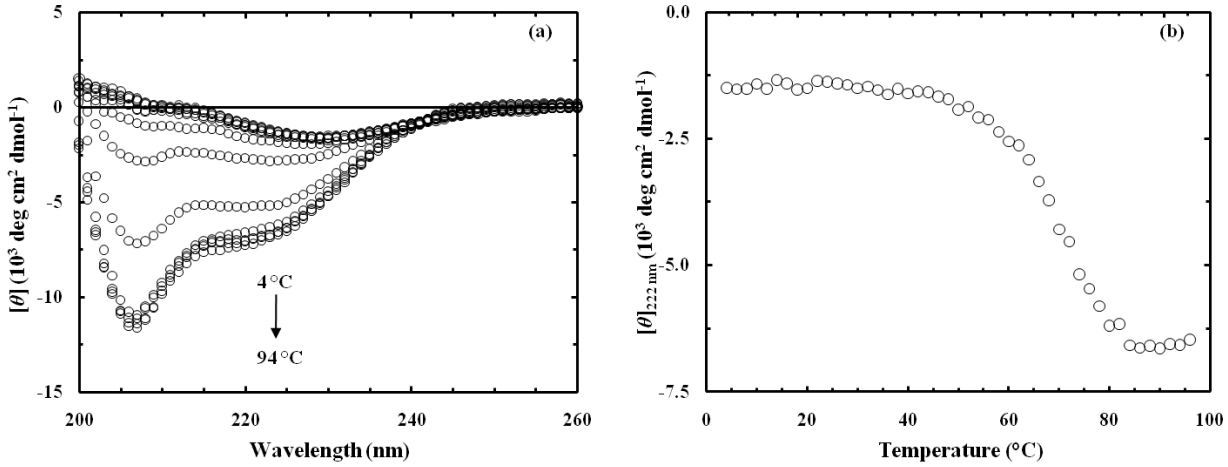

Figure 10. (a) Far-UV spectra and (b) mean residue ellipticity at 222 nm of AKA_2 in $[C_4mpy][Tf_2N]$ as a function of temperature. The spectra in (a) are shown for 10 °C intervals from 4 °C to 94 °C.

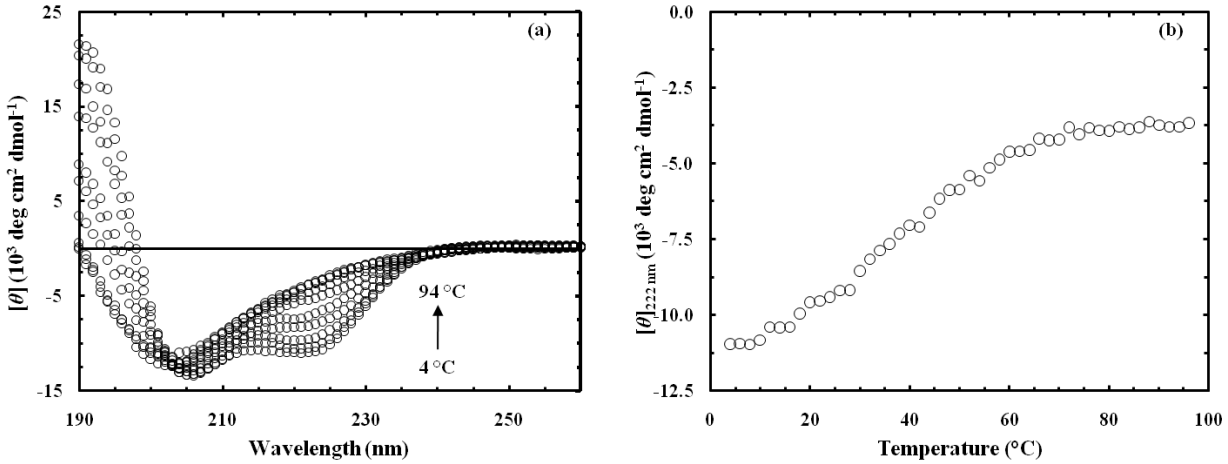

Figure 11. (a) Far-UV spectra and (b) mean residue ellipticity at 222 nm of Trp-cage in phosphate buffer as a function of temperature. The spectra in (a) are shown for 10 °C intervals from 4 °C to 94 °C.

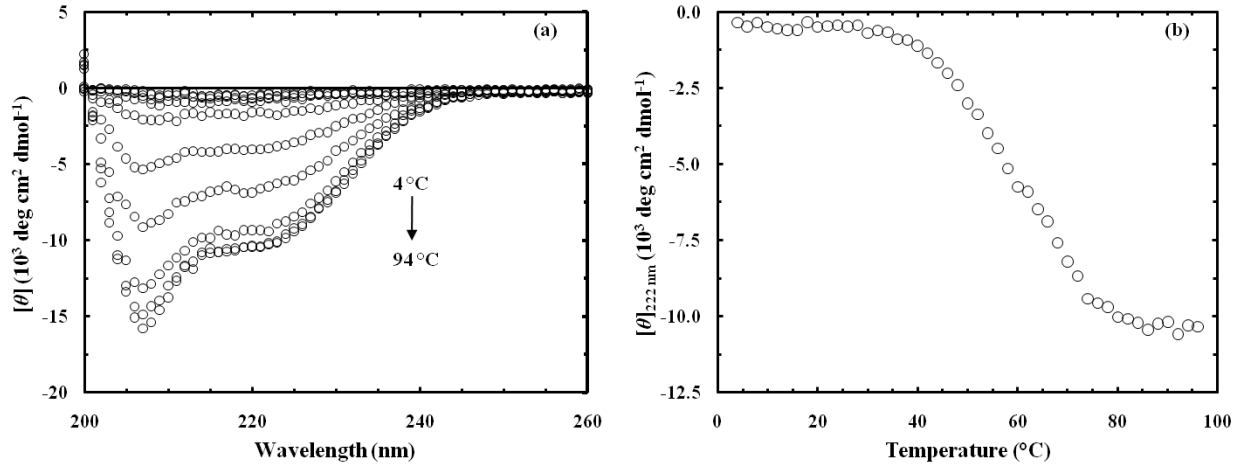

Figure 12. (a) Far-UV spectra and (b) mean residue ellipticity at 222 nm of Trp-cage in $[C_4mpy][Tf_2N]$ as a function of temperature. The spectra in (a) are shown for 10 °C intervals from 4 °C to 94 °C.

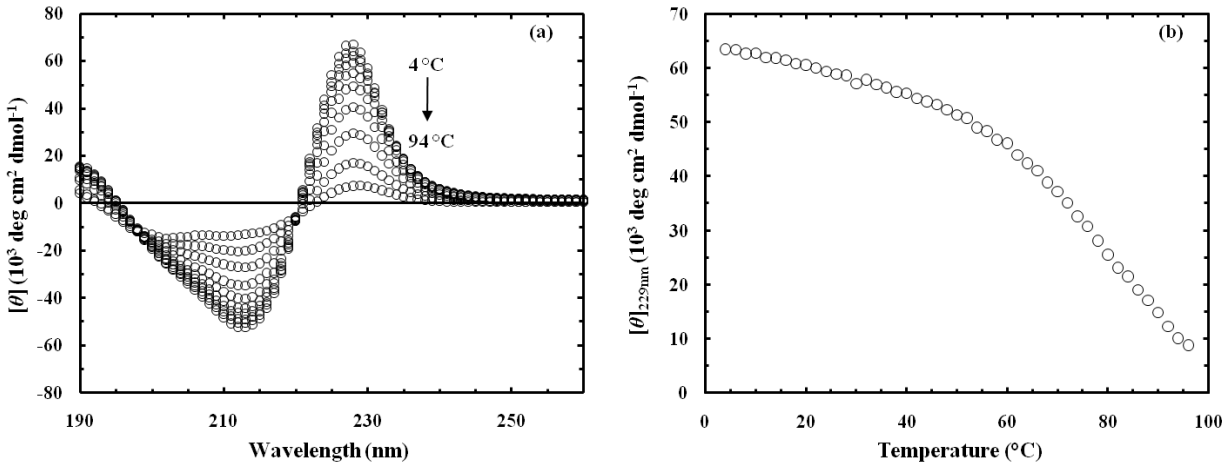

Figure 13. (a) Far-UV spectra and (b) mean residue ellipticity at 229 nm of Trpzip4 in phosphate buffer as a function of temperature. The spectra in (a) are shown for 10 °C intervals from 4 °C to 94 °C.

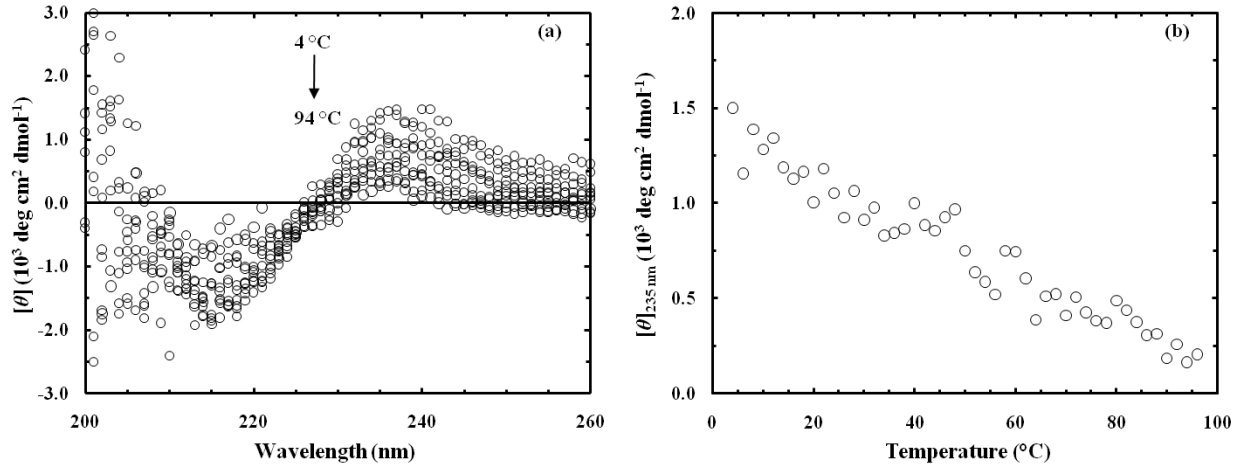

Figure 14. (a) Far-UV spectra and (b) mean residue ellipticity at 235 nm of Trpzip4 in $[C_4mpy][Tf_2N]$ as a function of temperature. The spectra in (a) are shown for 10 °C intervals from 4 °C to 94 °C.

References

1. S. M. Kelly, T. J. Jess and N. C. Price, *Biochim. Biophys. Acta*, 2005, **1751**, 119.