

Supplementary Information for

Structural Design and Functional Characterization of Dicationic Pyrazolium Salts as Organic Ionic Plastic Crystals

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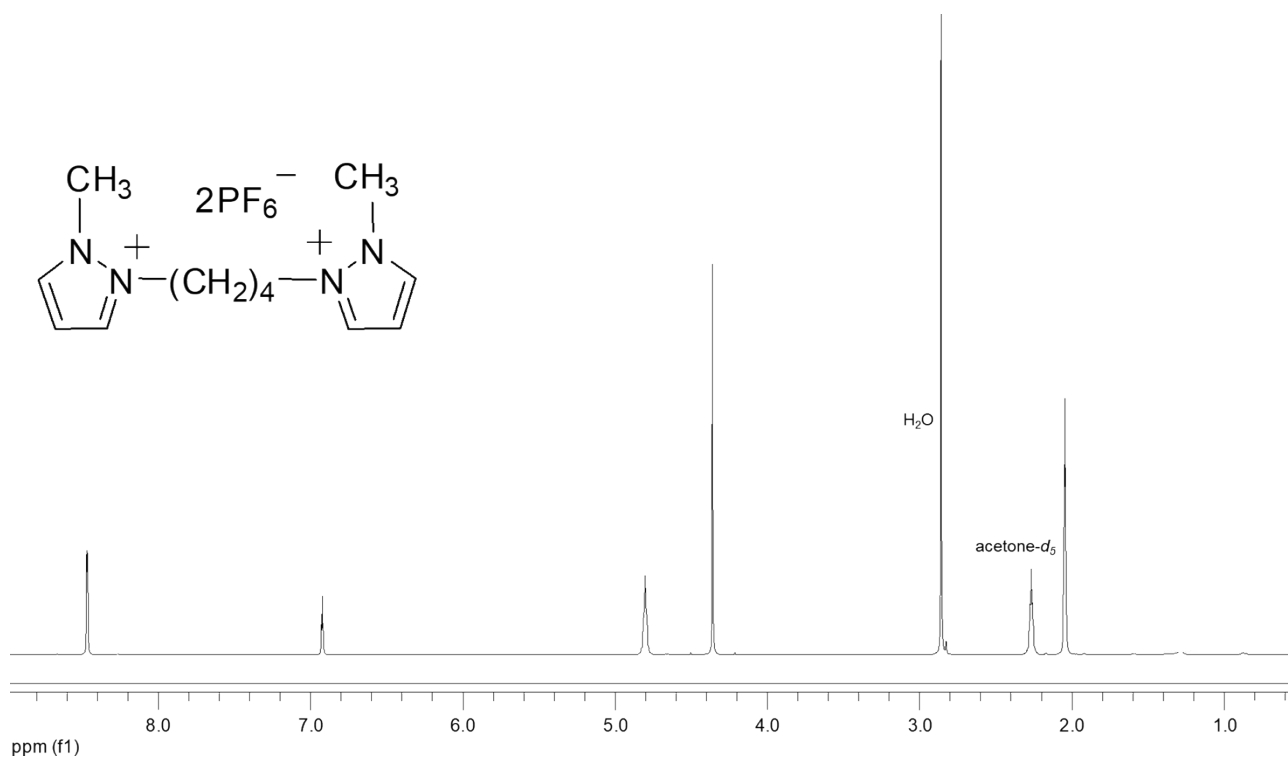


Figure S1. ^1H NMR spectrum of 1,2-bis[N-(N'-methylpyrazolium)butane] PF_6^- (**1-PF₆**) (acetone- d_6 , 23 °C)

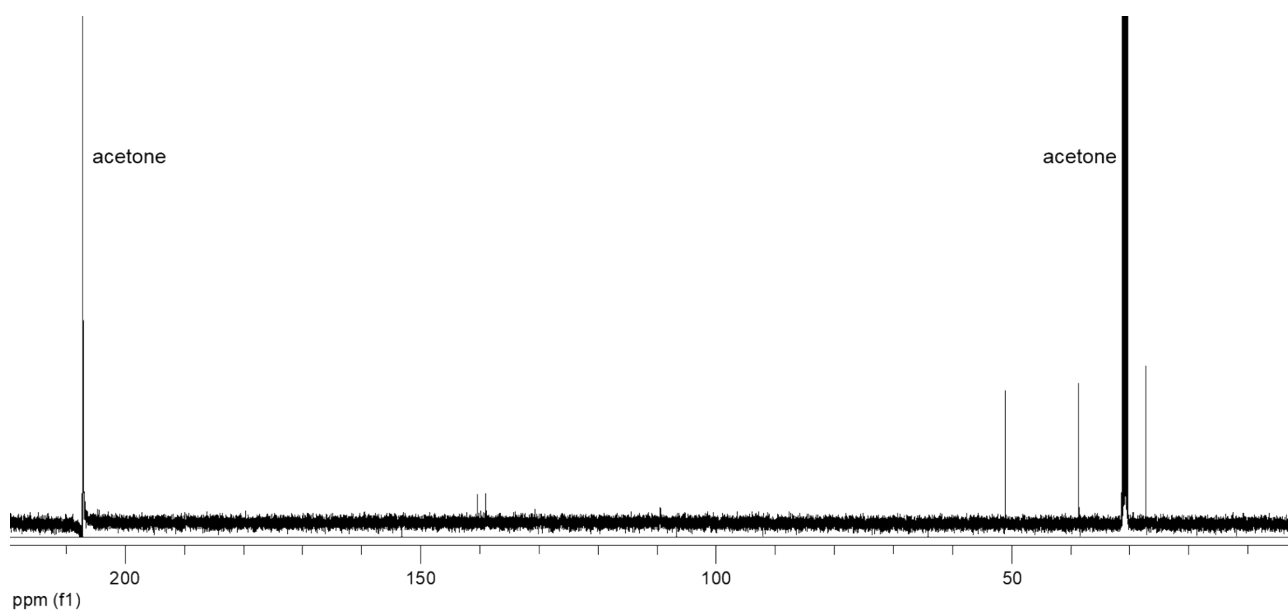


Figure S2. ^{13}C NMR spectrum of 1,2-bis[N-(N'-methylpyrazolium)butane] PF_6^- (**1-PF₆**) (acetone- d_6 , 23 °C)

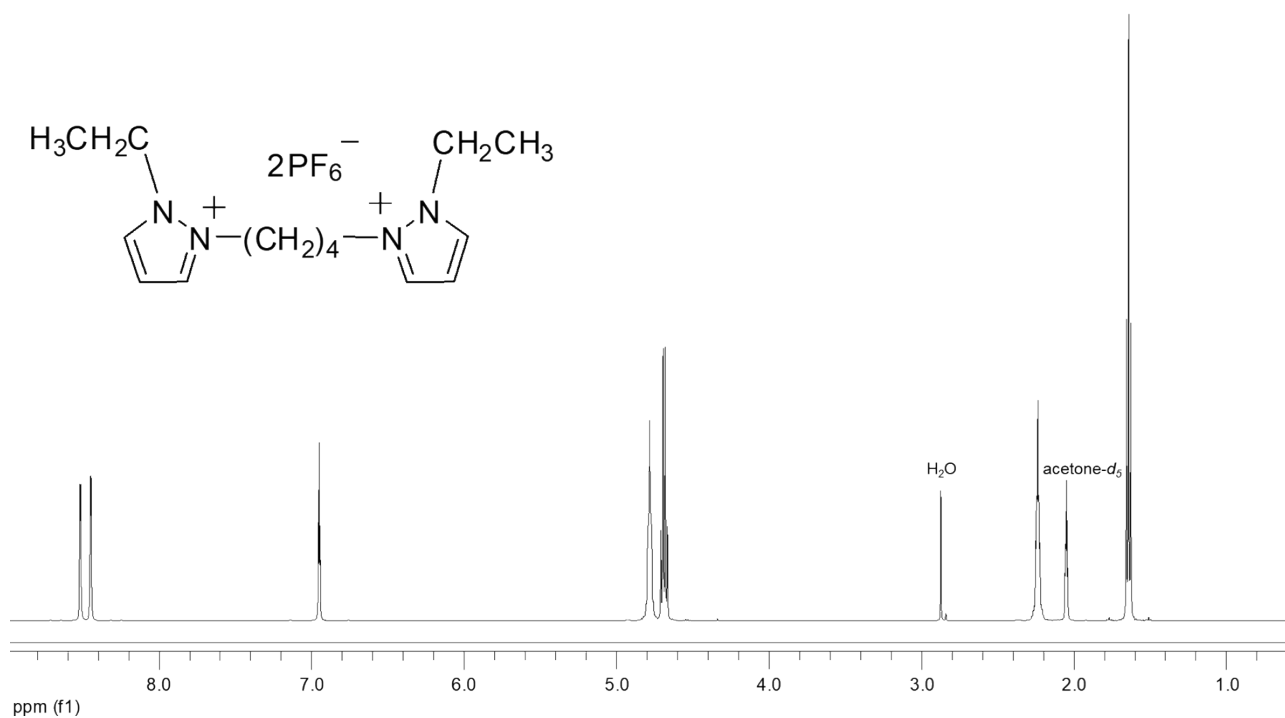


Figure S3. ¹H NMR spectrum of 1,2-bis[*N*-(*N'*-ethylpyrazolium)butane PF₆⁻ (**2-PF₆**) (acetone-*d*₆, 23 °C)

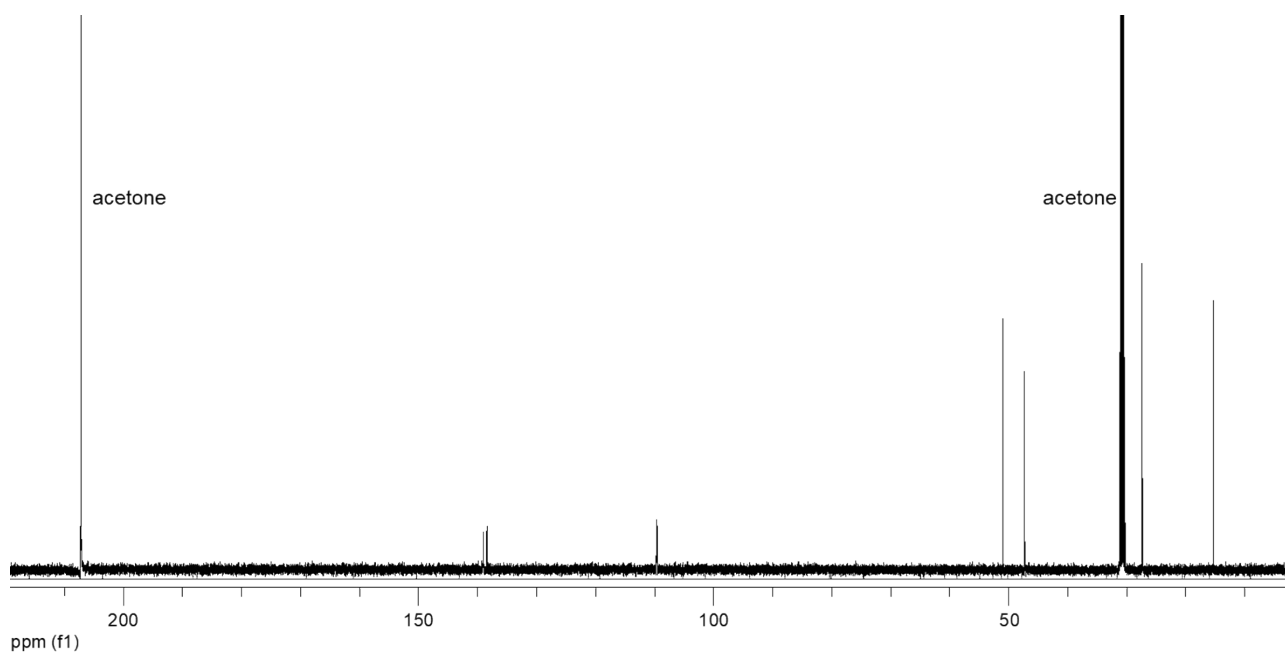


Figure S4. ¹³C NMR spectrum of 1,2-bis[*N*-(*N'*-ethylpyrazolium)butane PF₆⁻ (**2-PF₆**) (acetone-*d*₆, 23 °C)

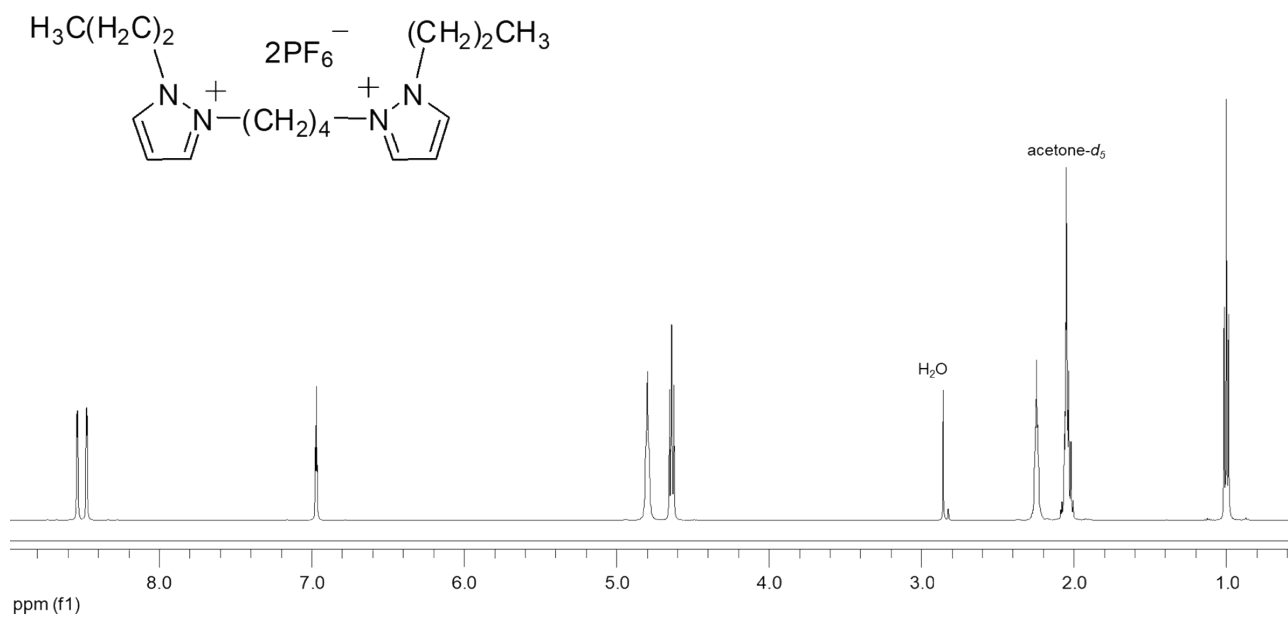


Figure S5. ¹H NMR spectrum of 1,2-bis[*N*-(*N*'-propylpyrazolium)butane] PF₆⁻ (**3-PF₆**) (acetone-*d*₆, 23 °C)

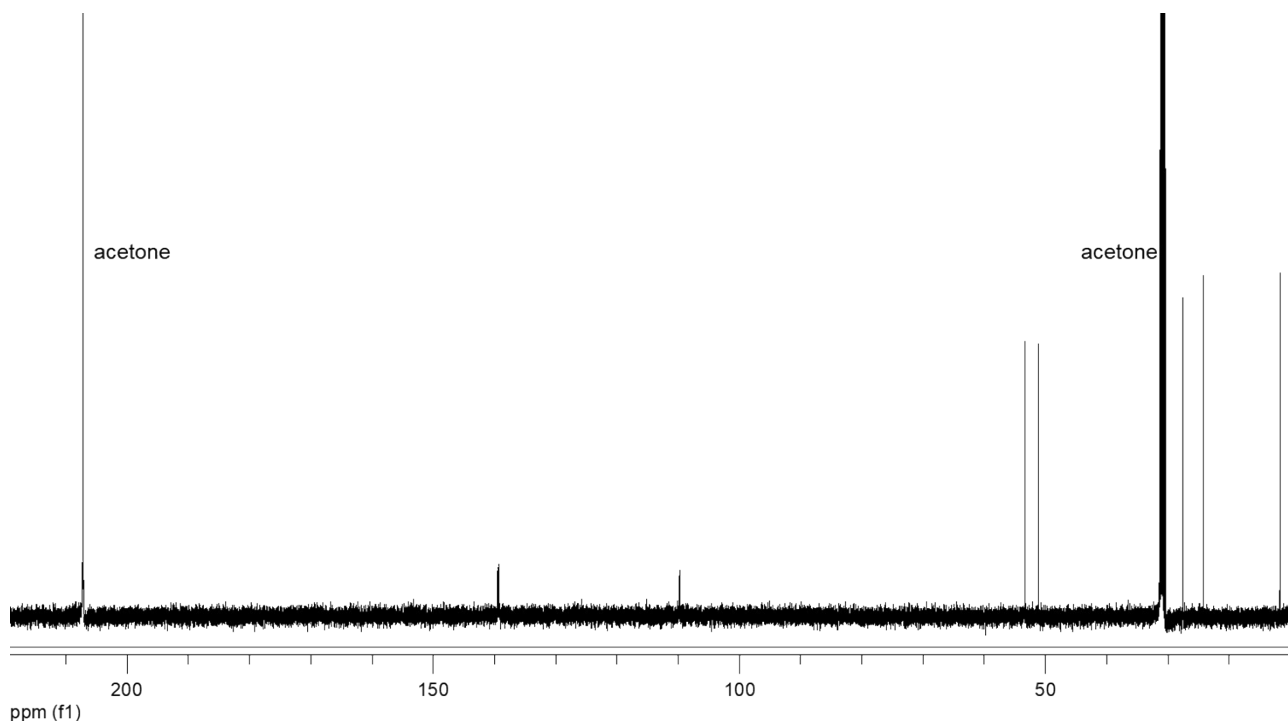


Figure S6. ¹³C NMR spectrum of 1,2-bis[*N*-(*N*'-propylpyrazolium)butane] PF₆⁻ (**3-PF₆**) (acetone-*d*₆, 23 °C)

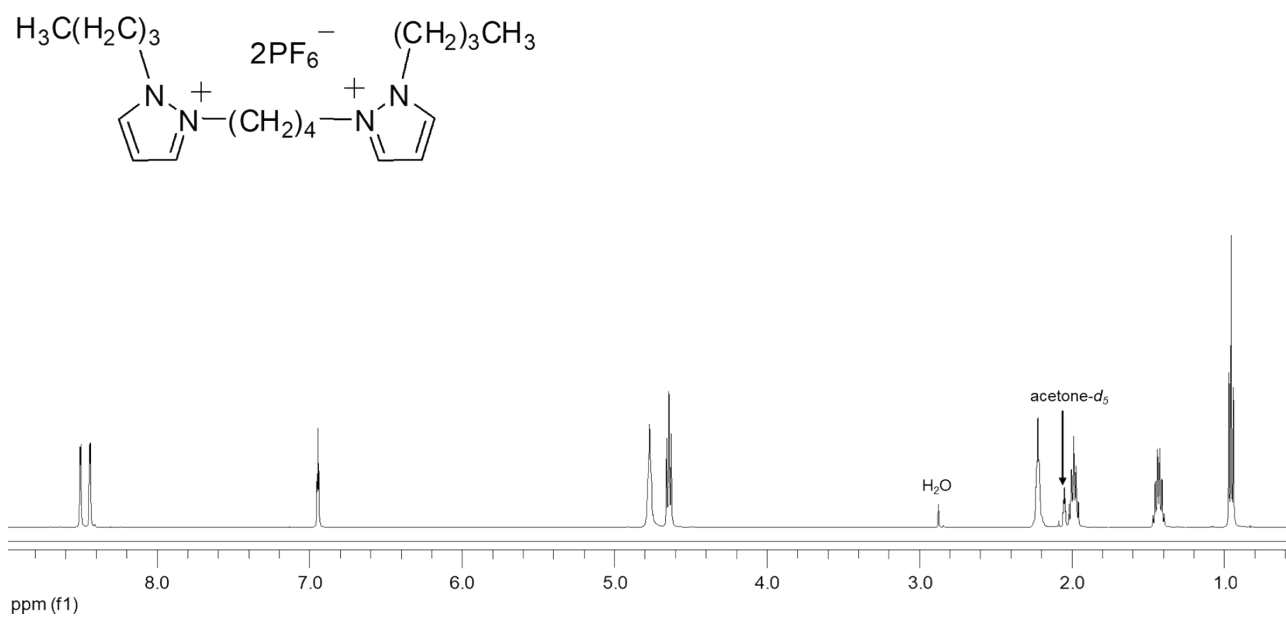


Figure S7. ¹H NMR spectrum of 1,2-bis[*N*-(*N'*-butylpyrazolium)butane] PF₆⁻ (**4-PF₆**) (acetone-*d*₆, 23 °C)

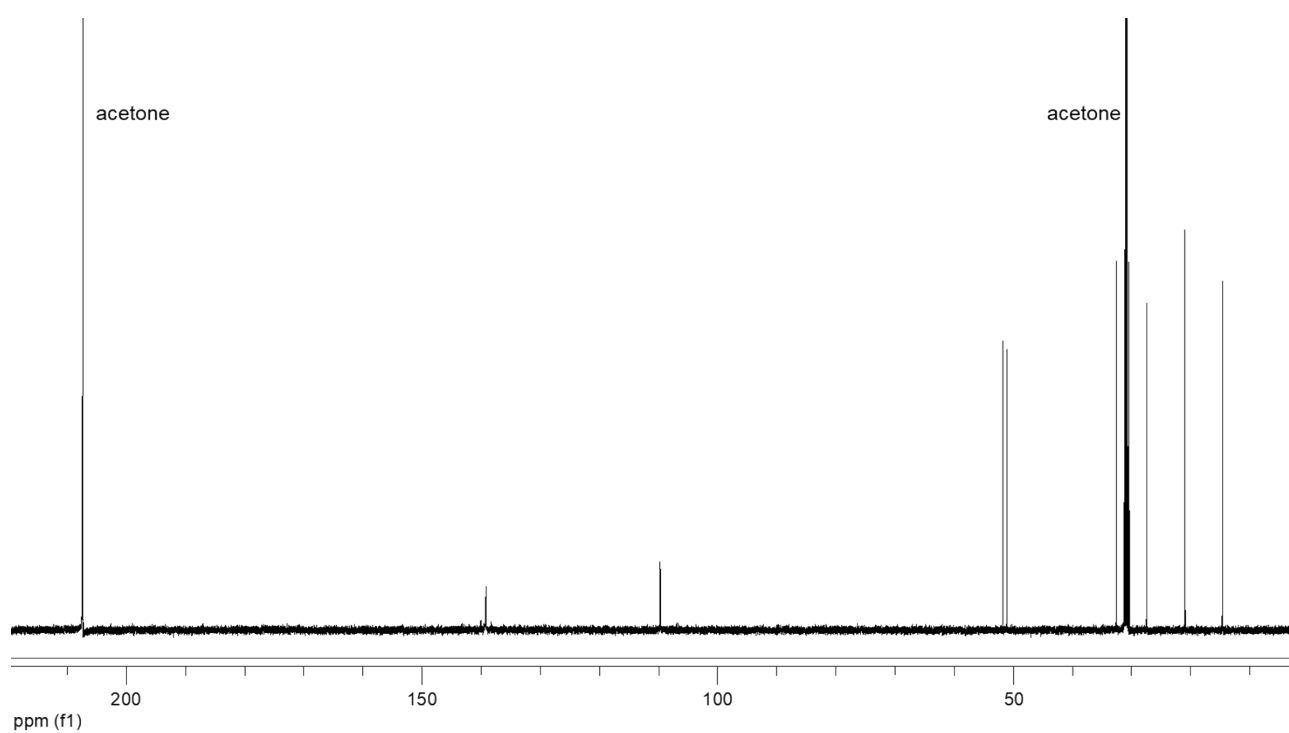


Figure S8. ¹³C NMR spectrum of 1,2-bis[*N*-(*N'*-butylpyrazolium)butane] PF₆⁻ (**4-PF₆**) (acetone-*d*₆, 23 °C)

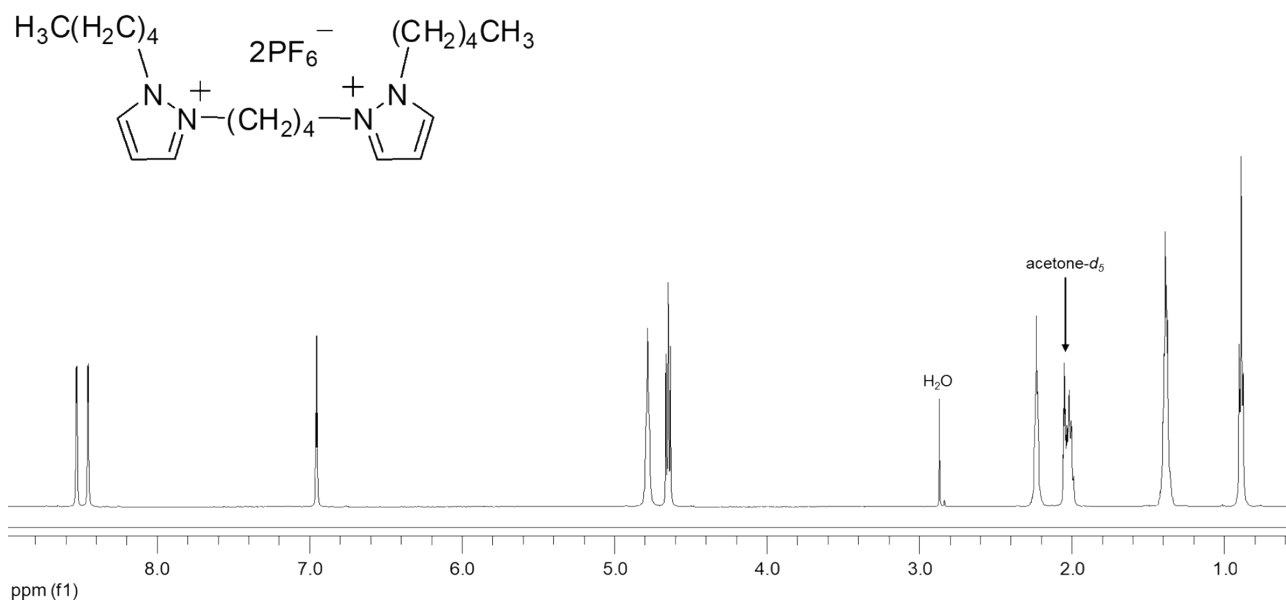


Figure S9. ¹H NMR spectrum of 1,2-bis[*N*-(*N*'-pentylpyrazolium)butane] PF₆⁻ (**5-PF₆**) (acetone-*d*₆, 23 °C)

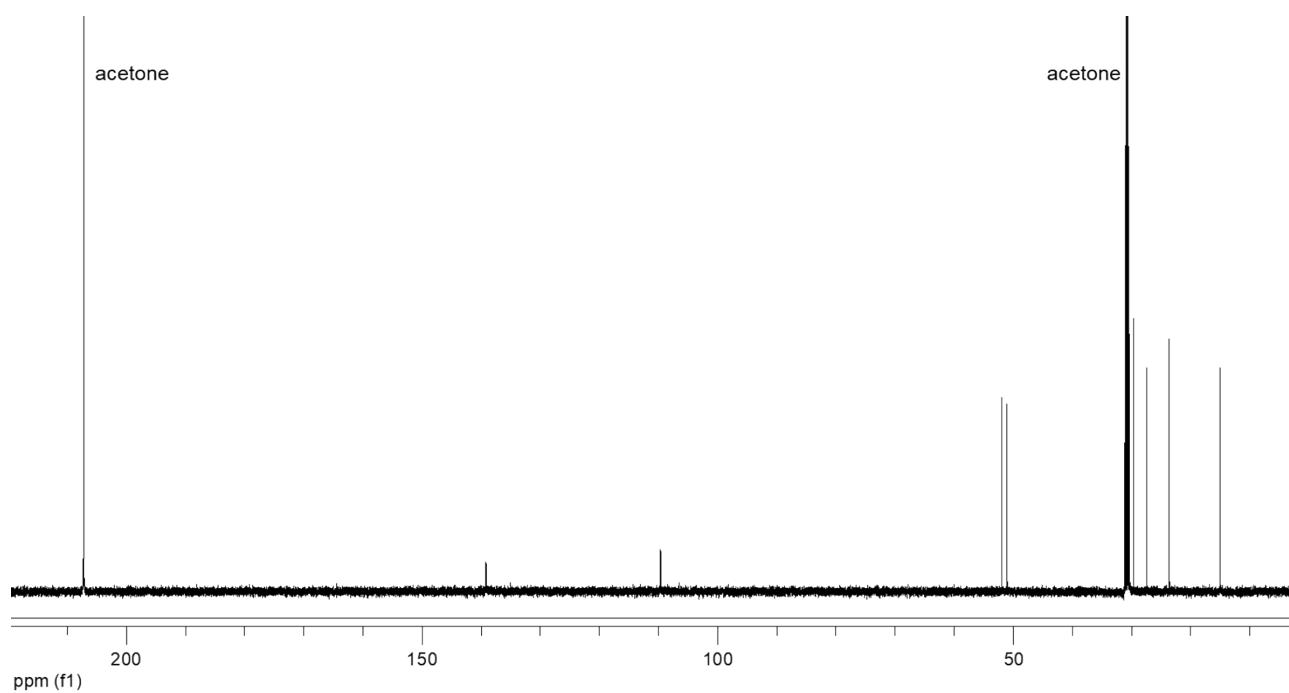


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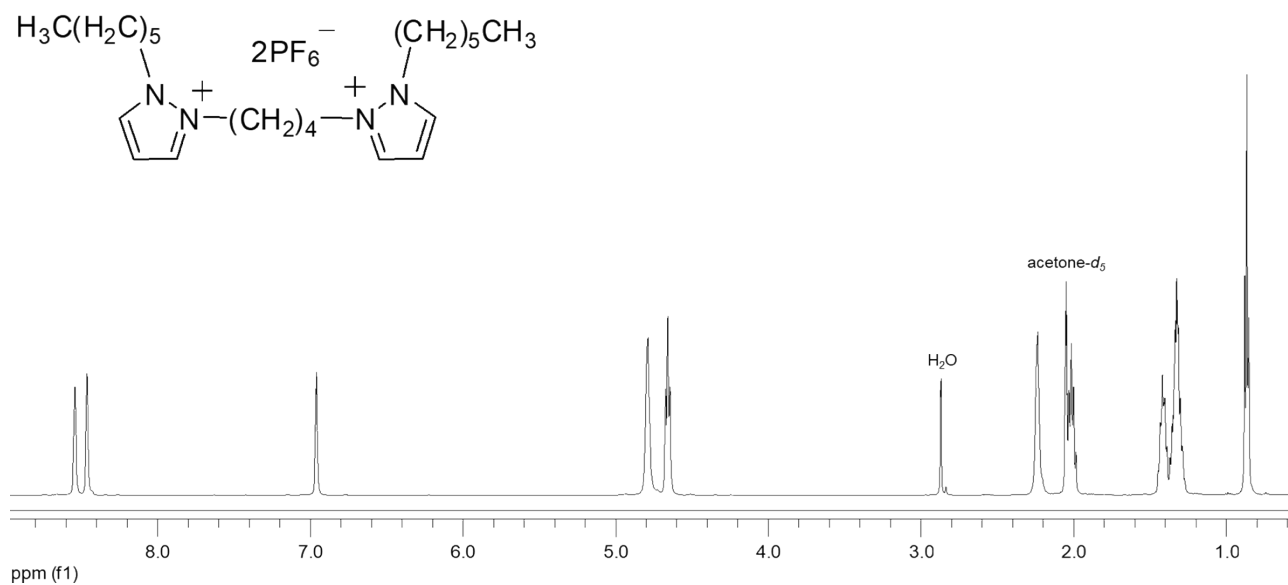


Figure S11. ¹H NMR spectrum of 1,2-bis[*N*-(*N'*-hexylpyrazolium)butane] PF₆⁻ (**6-PF₆**) (acetone-*d*₆, 23 °C)

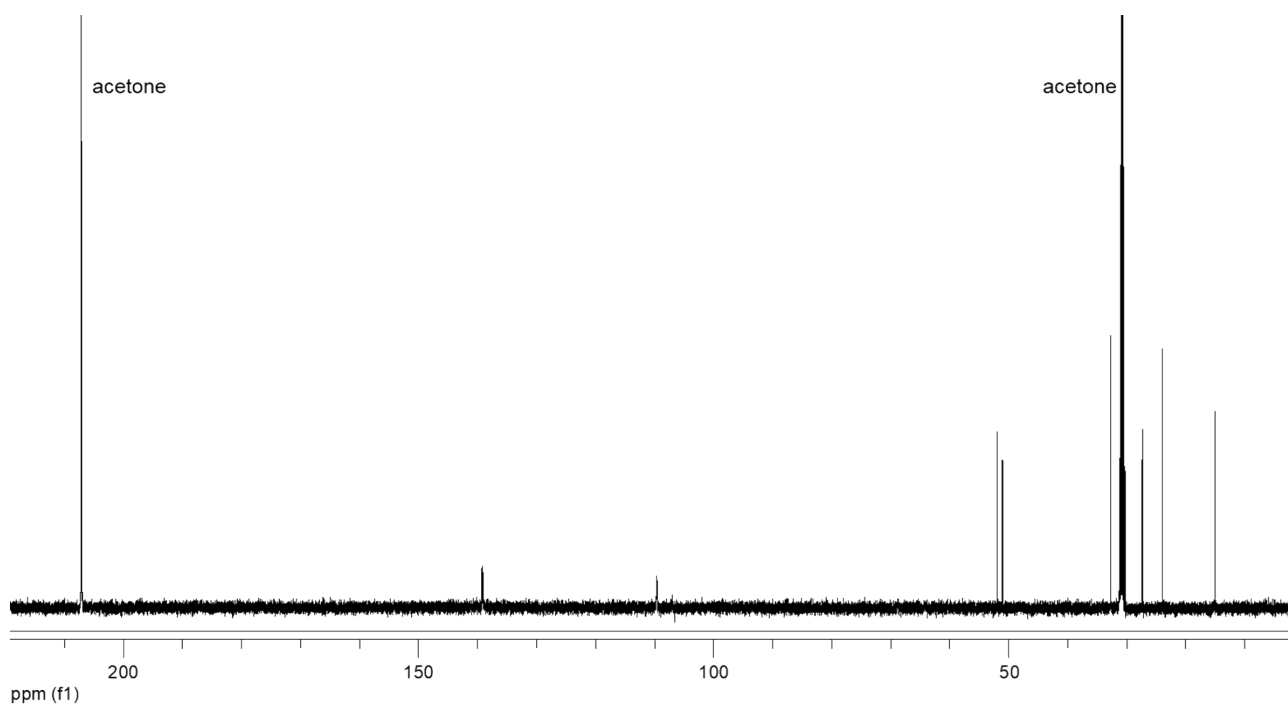


Figure S12. ¹³C NMR spectrum of 1,2-bis[*N*-(*N'*-hexylpyrazolium)butane] PF₆⁻ (**6-PF₆**) (acetone-*d*₆, 23 °C)

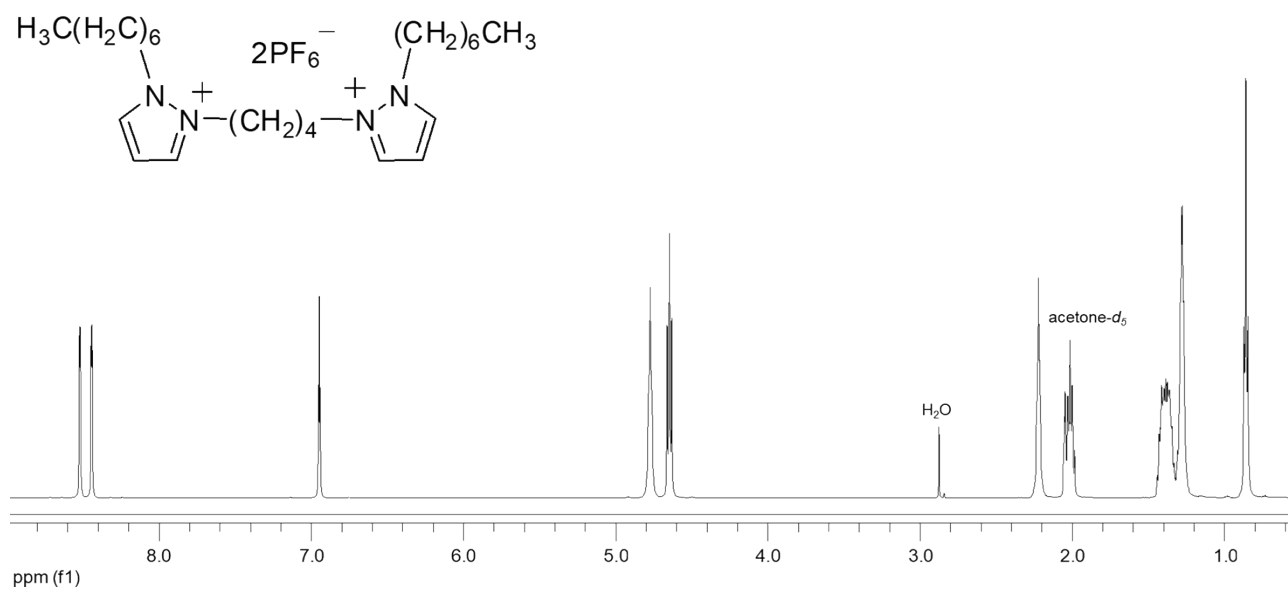


Figure S13. ¹H NMR spectrum of 1,2-bis[*N*-(*N'*-heptylpyrazolium)butane] PF₆⁻ (**7-PF₆**) (acetone-*d*₆, 23 °C)

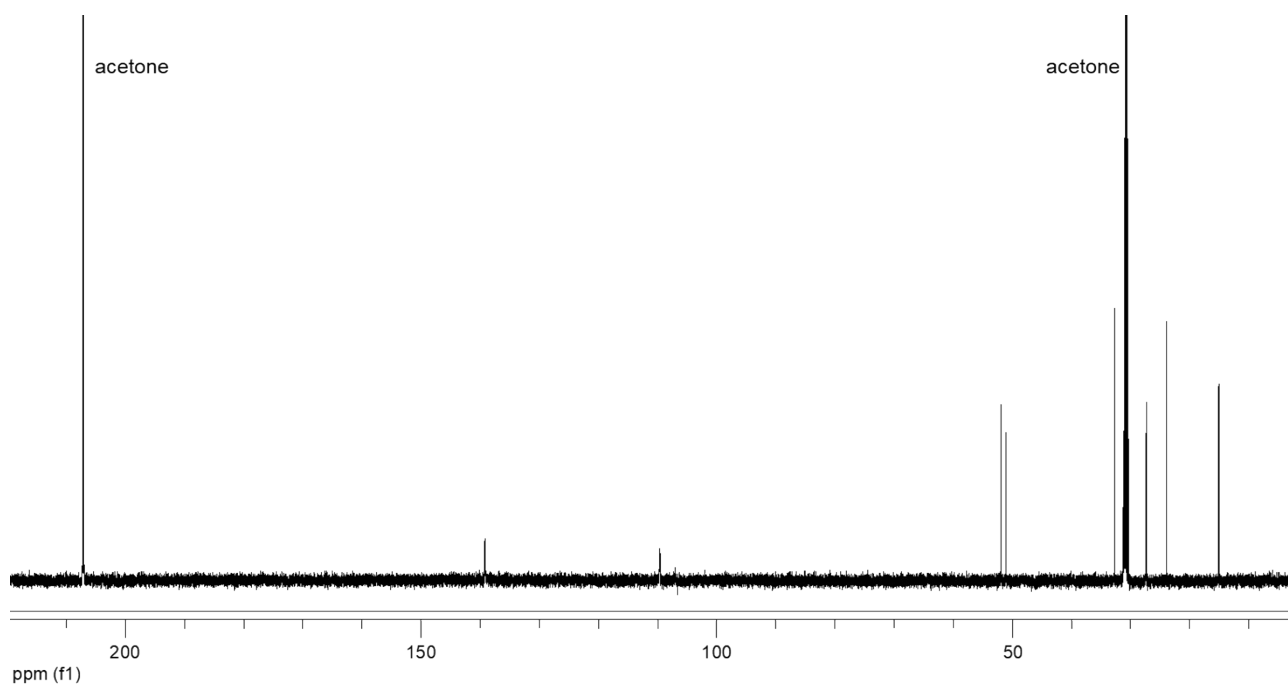


Figure S14. ¹³C NMR spectrum of 1,2-bis[*N*-(*N'*-heptylpyrazolium)butane] PF₆⁻ (**7-PF₆**) (acetone-*d*₆, 23 °C)

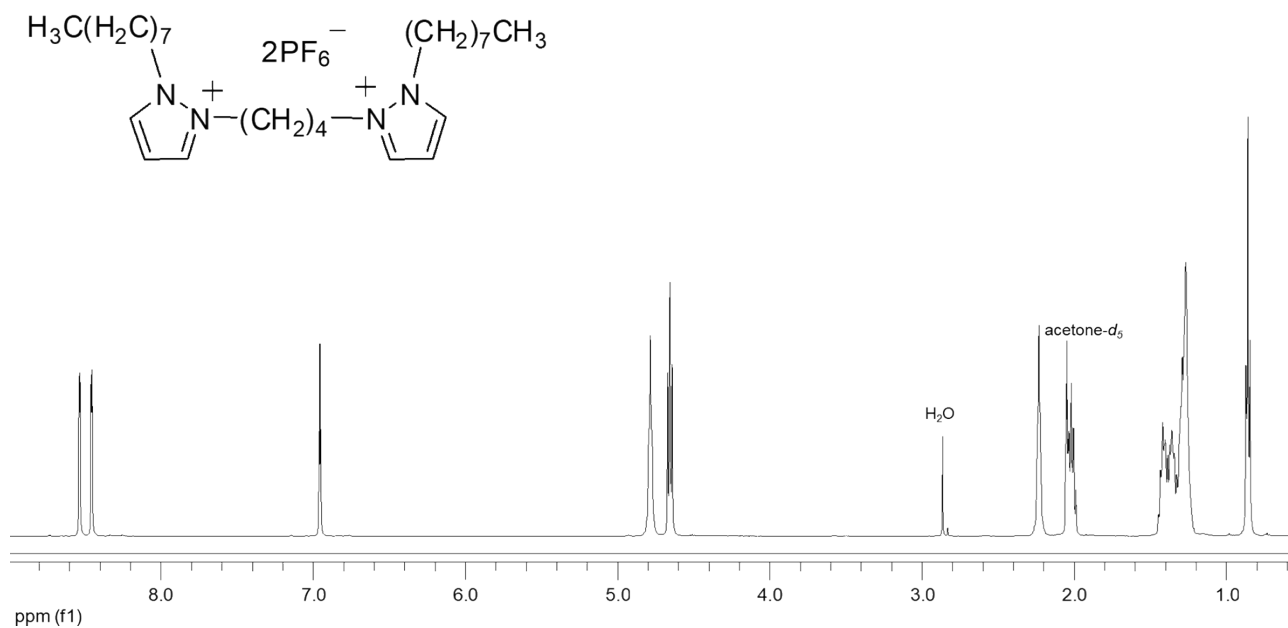


Figure S15. ¹H NMR spectrum of 1,2-bis[*N*-(*N'*-octylpyrazolium)butane] PF₆⁻ (**8-PF₆**) (acetone-*d*₆, 23 °C)

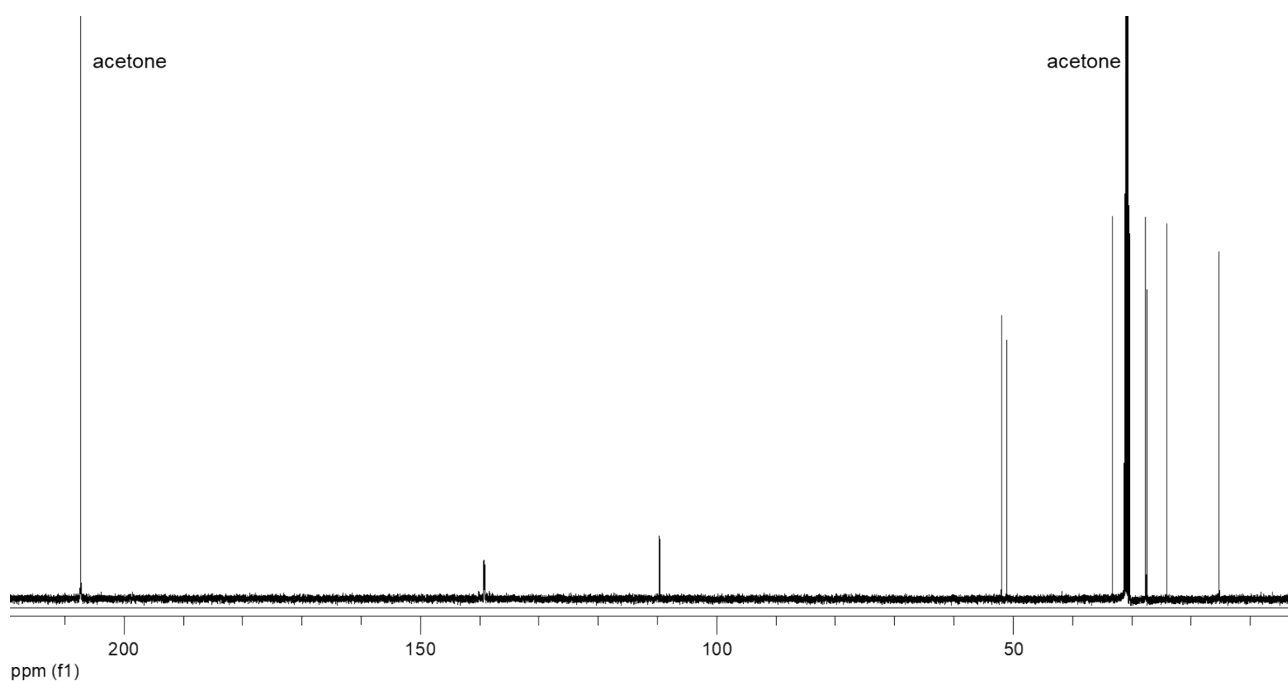


Figure S16. ¹³C NMR spectrum of 1,2-bis[*N*-(*N'*-octylpyrazolium)butane] PF₆⁻ (**8-PF₆**) (acetone-*d*₆, 23 °C)

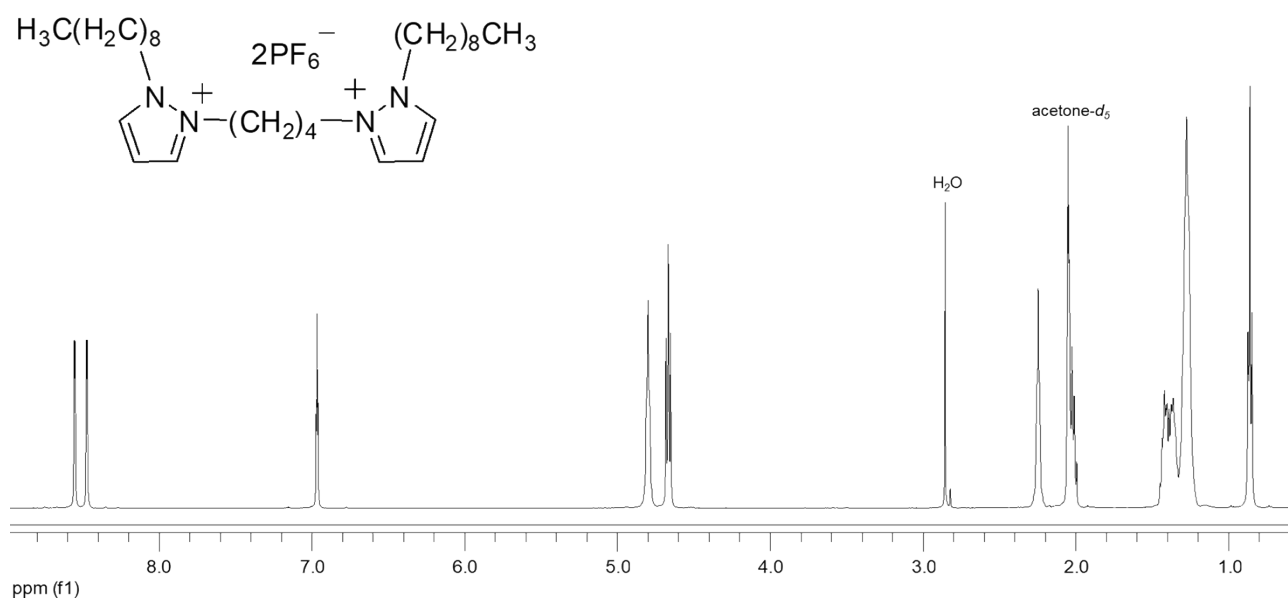


Figure S17. ¹H NMR spectrum of 1,2-bis[*N*-(*N'*-nonylpyrazolium)butane] PF₆⁻ (**9-PF₆**) (Acetone-*d*₆, 23 °C)

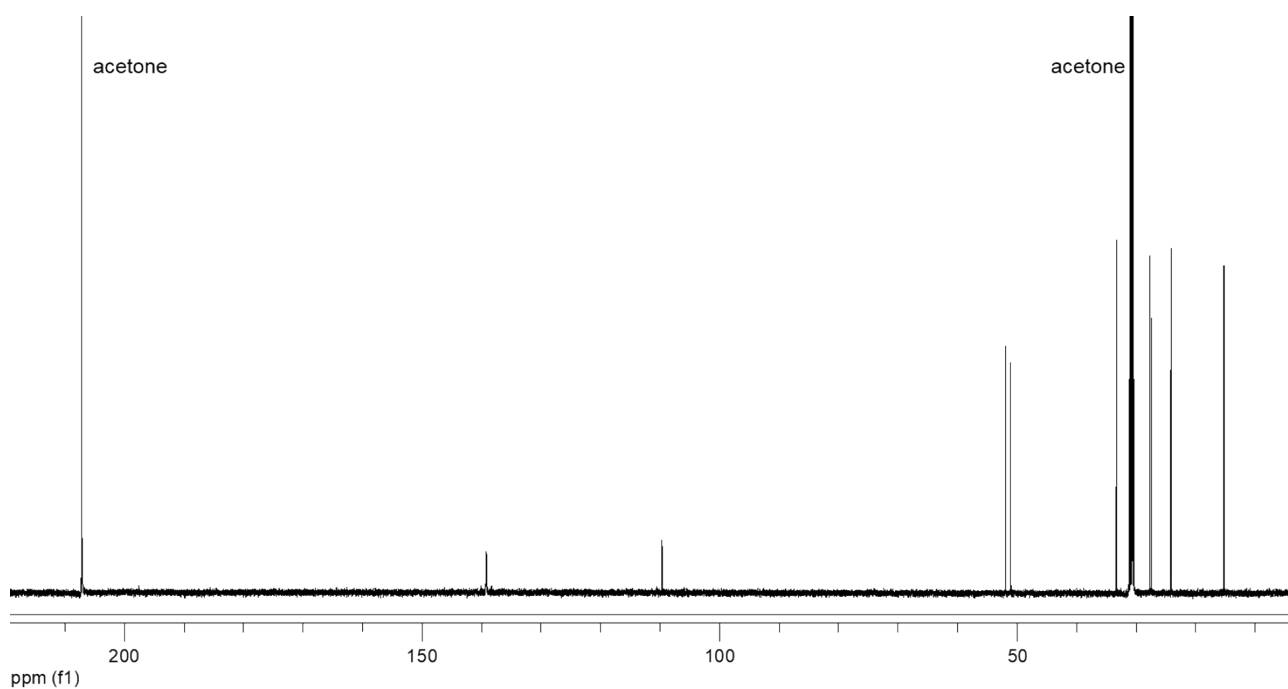


Figure S18. ¹³C NMR spectrum of 1,2-bis[*N*-(*N'*-nonylpyrazolium)butane] PF₆⁻ (**9-PF₆**) (acetone-*d*₆, 23 °C)

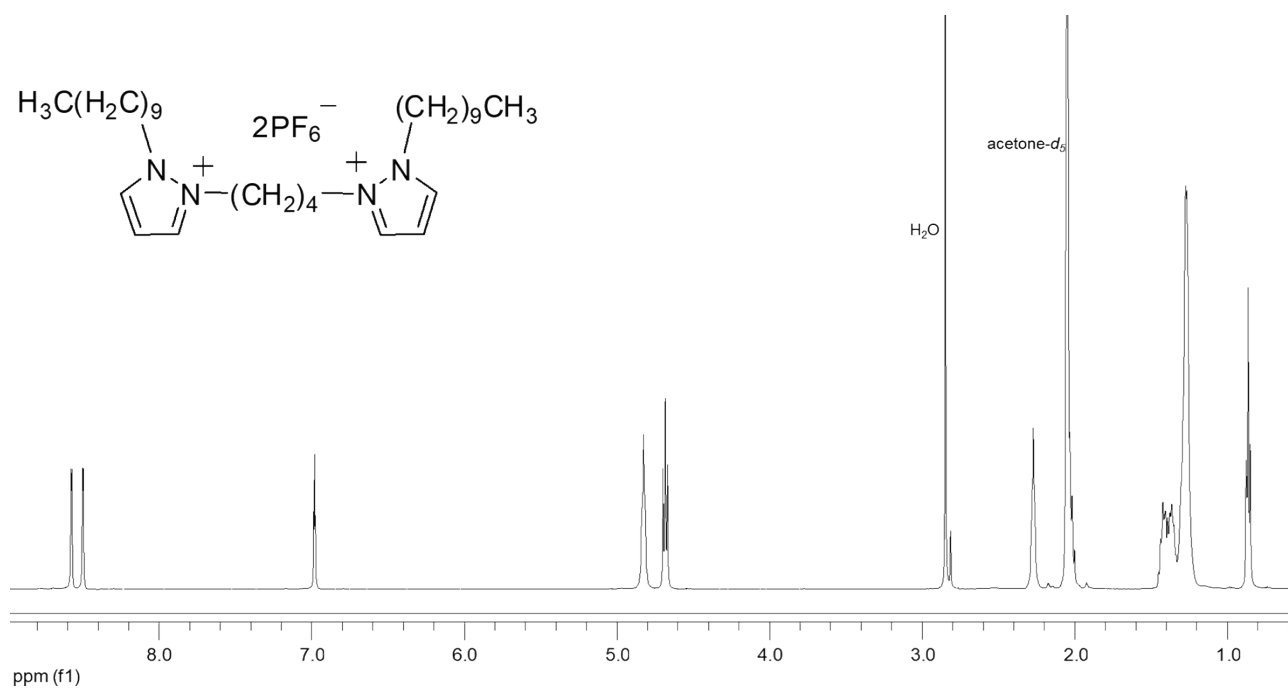


Figure S19. ¹H NMR spectrum of 1,2-bis[*N*-(*N'*-decylpyrazolium)butane] PF₆⁻ (**10-PF₆**) (Acetone-*d*₆, 23 °C)

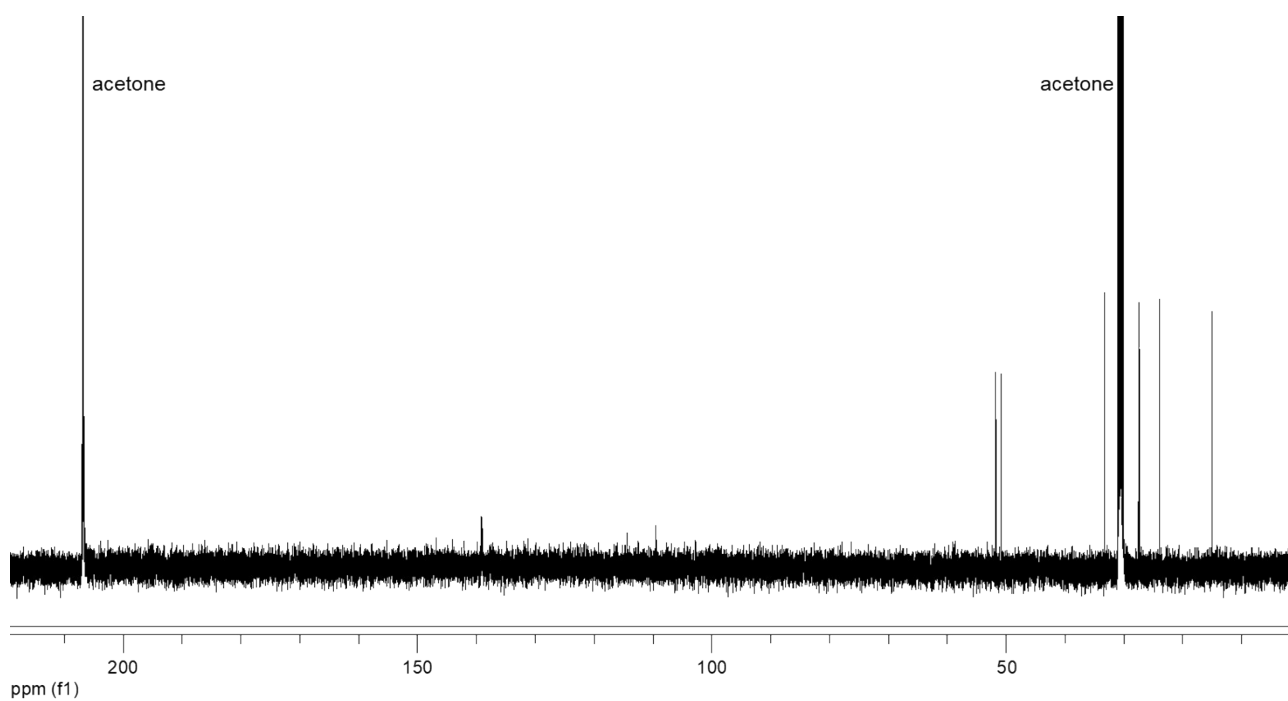


Figure S20. ¹³C NMR spectrum of 1,2-bis[*N*-(*N'*-decylpyrazolium)butane] PF₆⁻ (**10-PF₆**) (acetone-*d*₆, 23 °C)

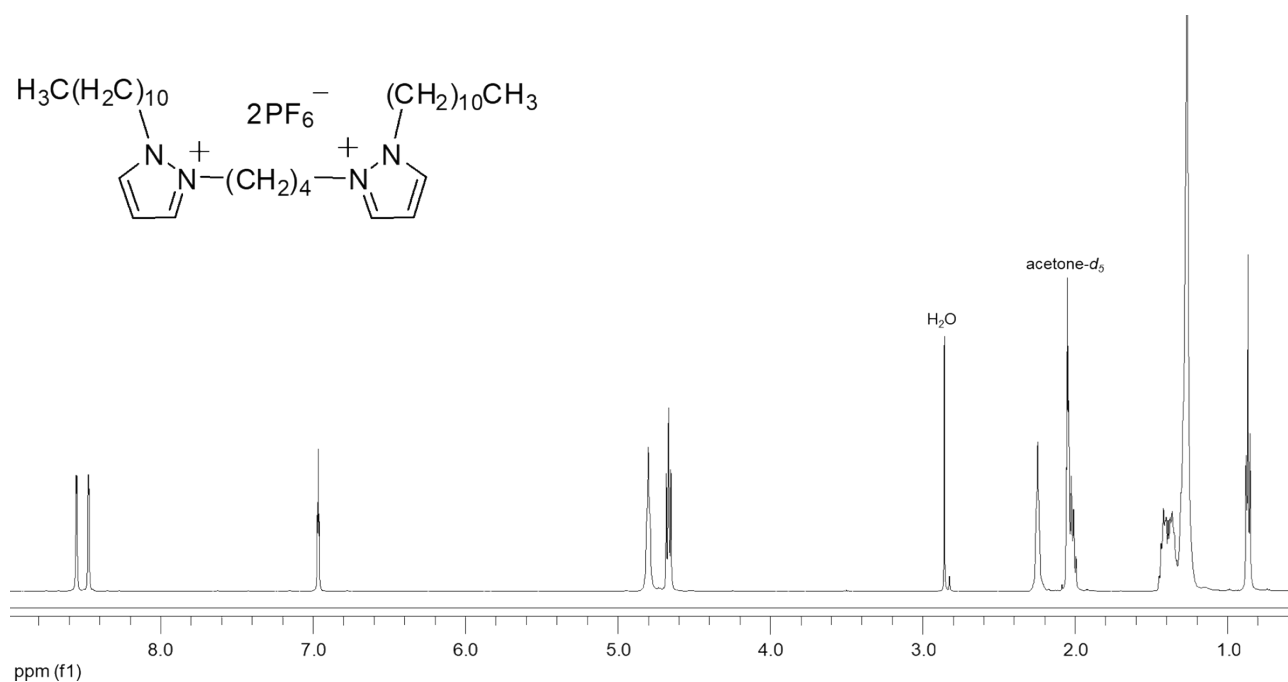


Figure S21. ¹H NMR spectrum of 1,2-bis[*N*-(*N'*-undecylpyrazolium)butane] PF₆⁻ (**11-PF₆**) (acetone-*d*₆, 23 °C)

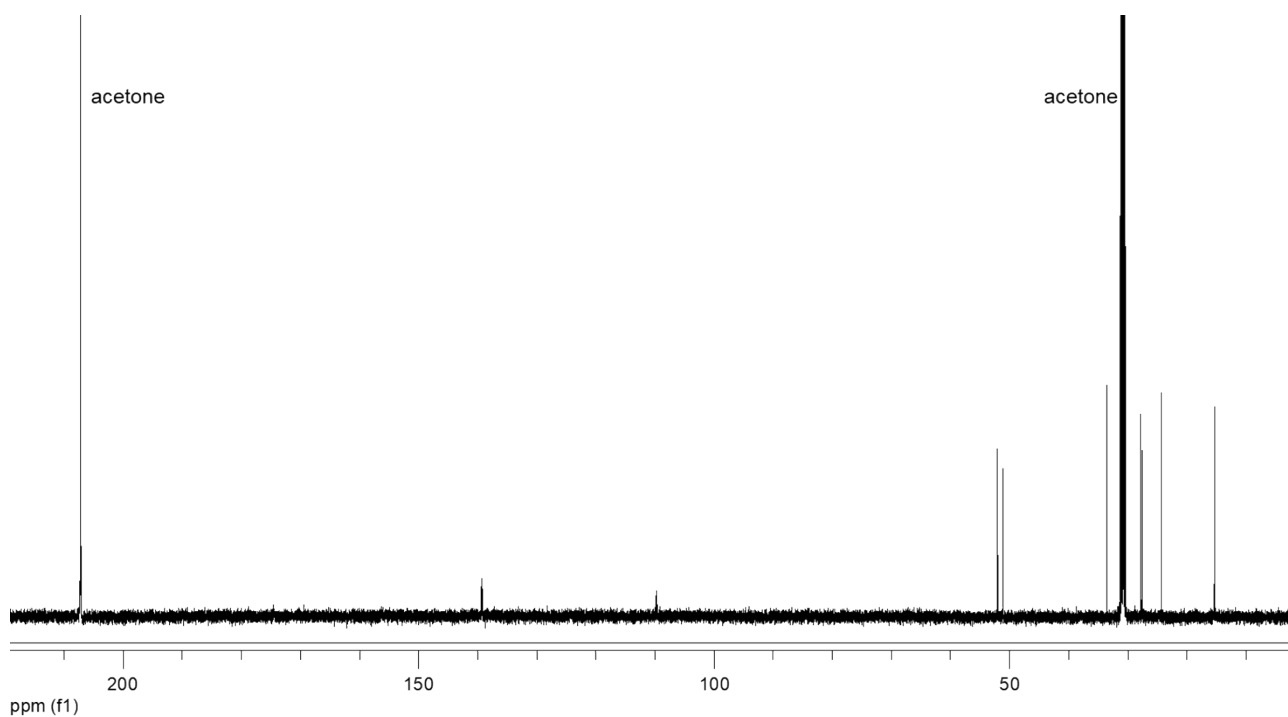


Figure S22. ¹³C NMR spectrum of 1,2-bis[*N*-(*N'*-undecylpyrazolium)butane] PF₆⁻ (**11-PF₆**) (acetone-*d*₆, 23 °C)

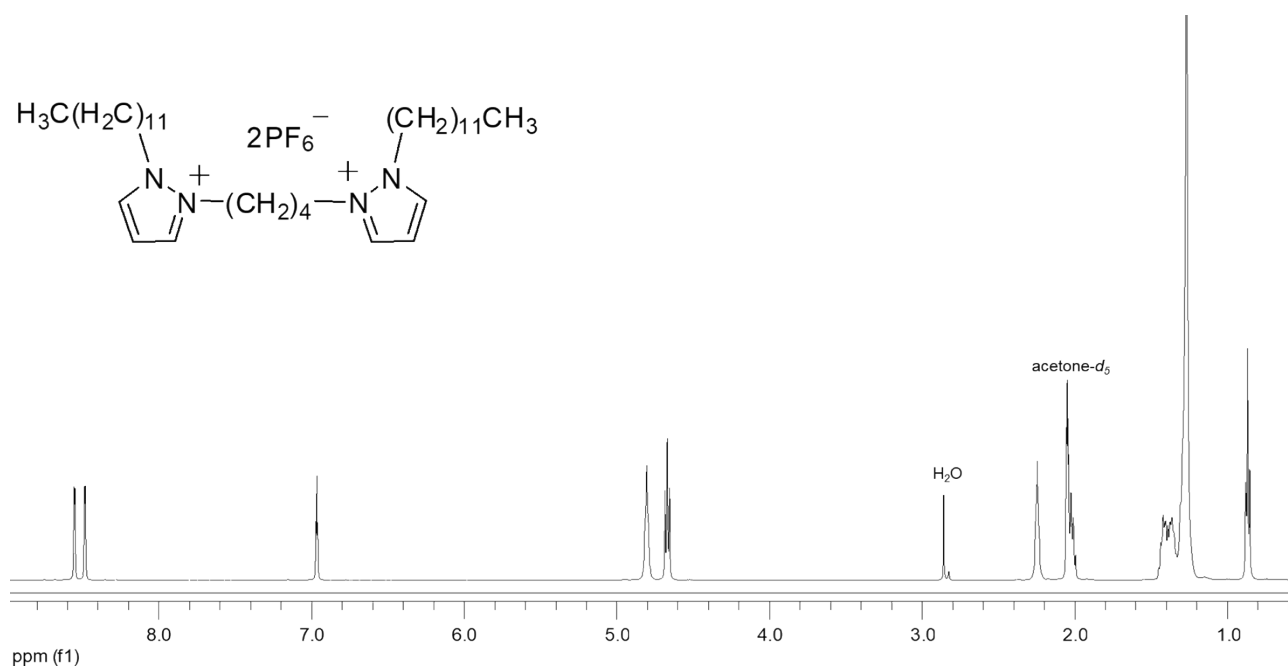


Figure S23. ¹H NMR spectrum of 1,2-bis[*N*-(*N'*-dodecylpyrazolium)butane] PF₆⁻ (**12-PF₆**) (acetone-*d*₆, 23 °C)

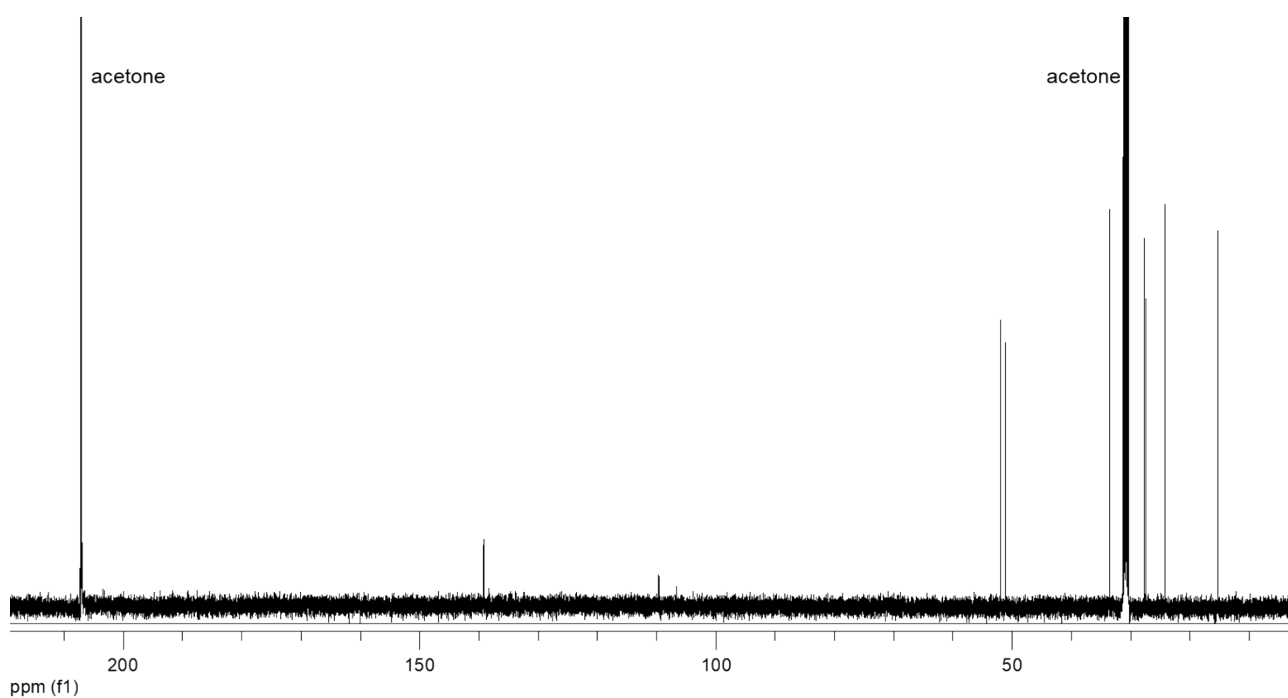


Figure S24. ¹³C NMR spectrum of 1,2-bis[*N*-(*N'*-dodecylpyrazolium)butane] PF₆⁻ (**12-PF₆**) (acetone-*d*₆, 23 °C)

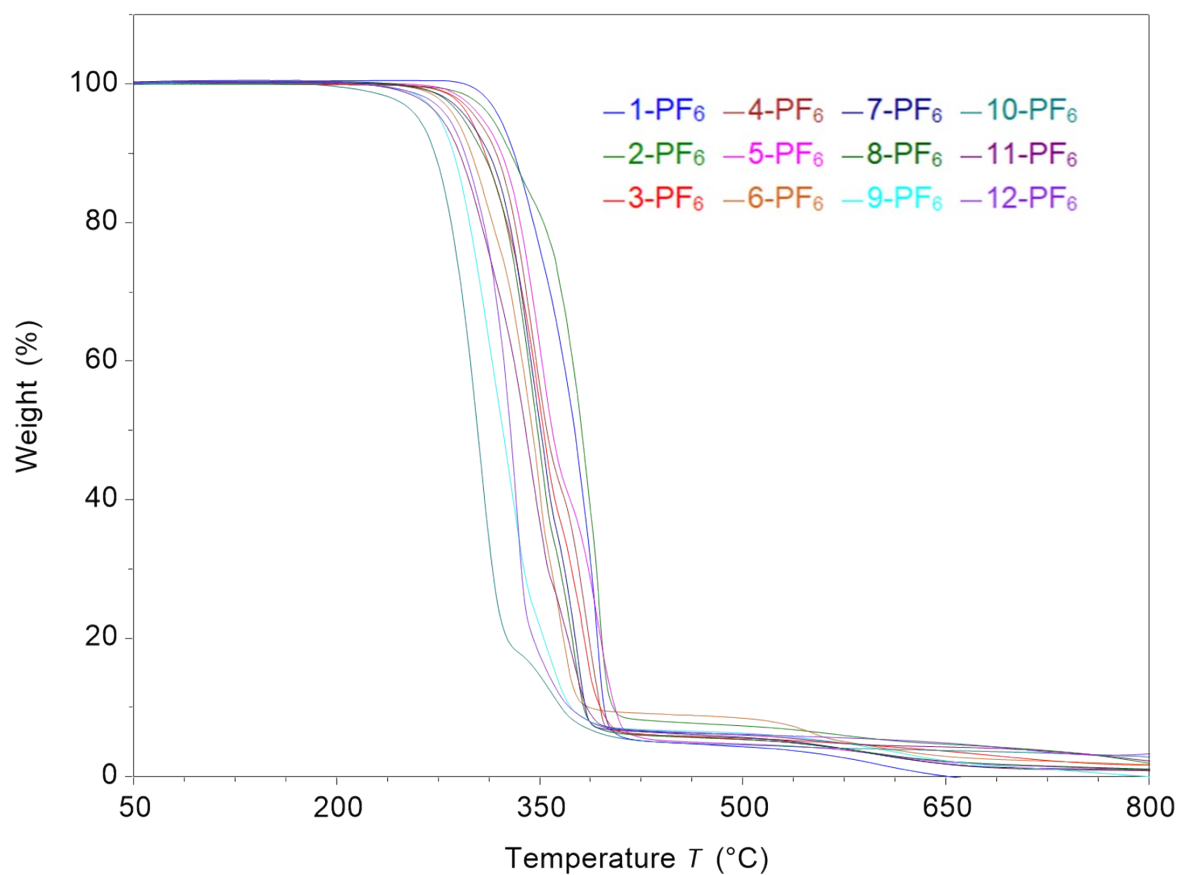


Figure S25. TGA thermograms of 1,2-bis[*N*-(*N'*-alkylpyrazolium)butane PF_6^-

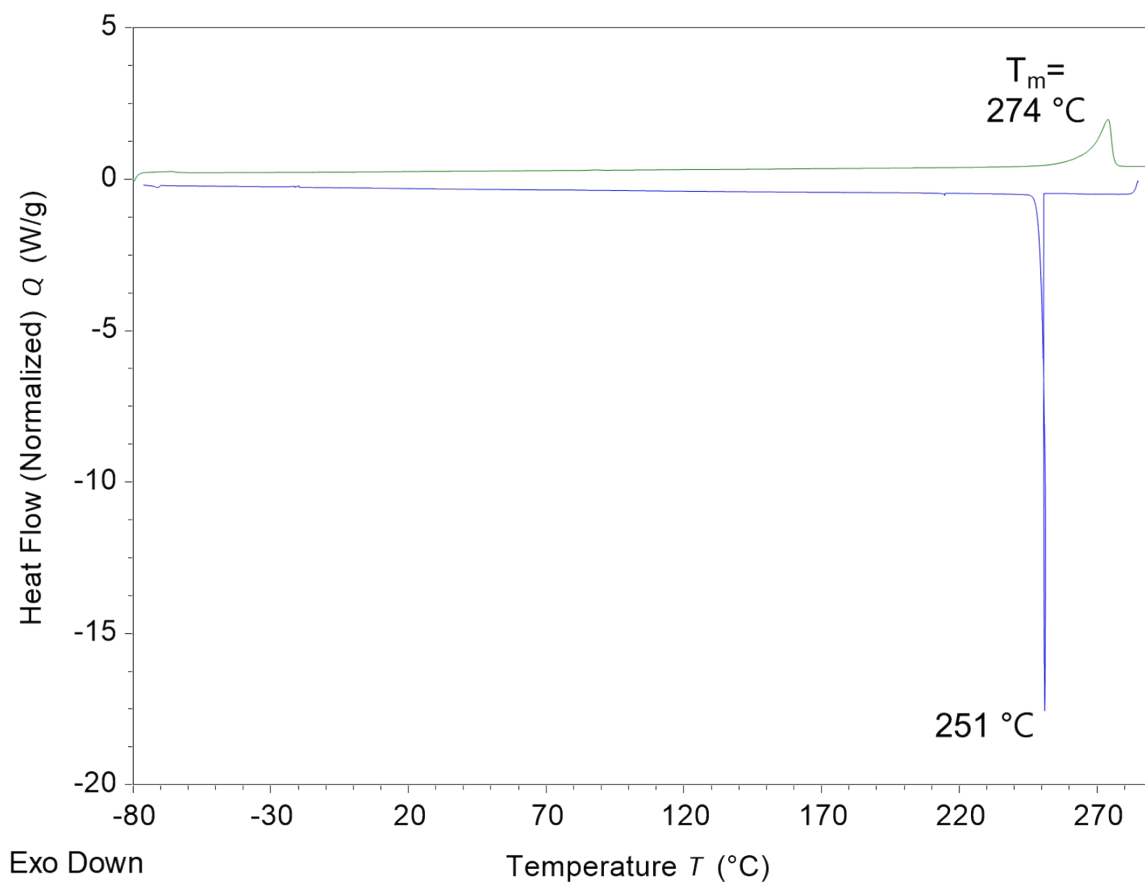


Figure S26. DSC 2nd heating (green) and cooling (blue) curves of **1-PF₆**

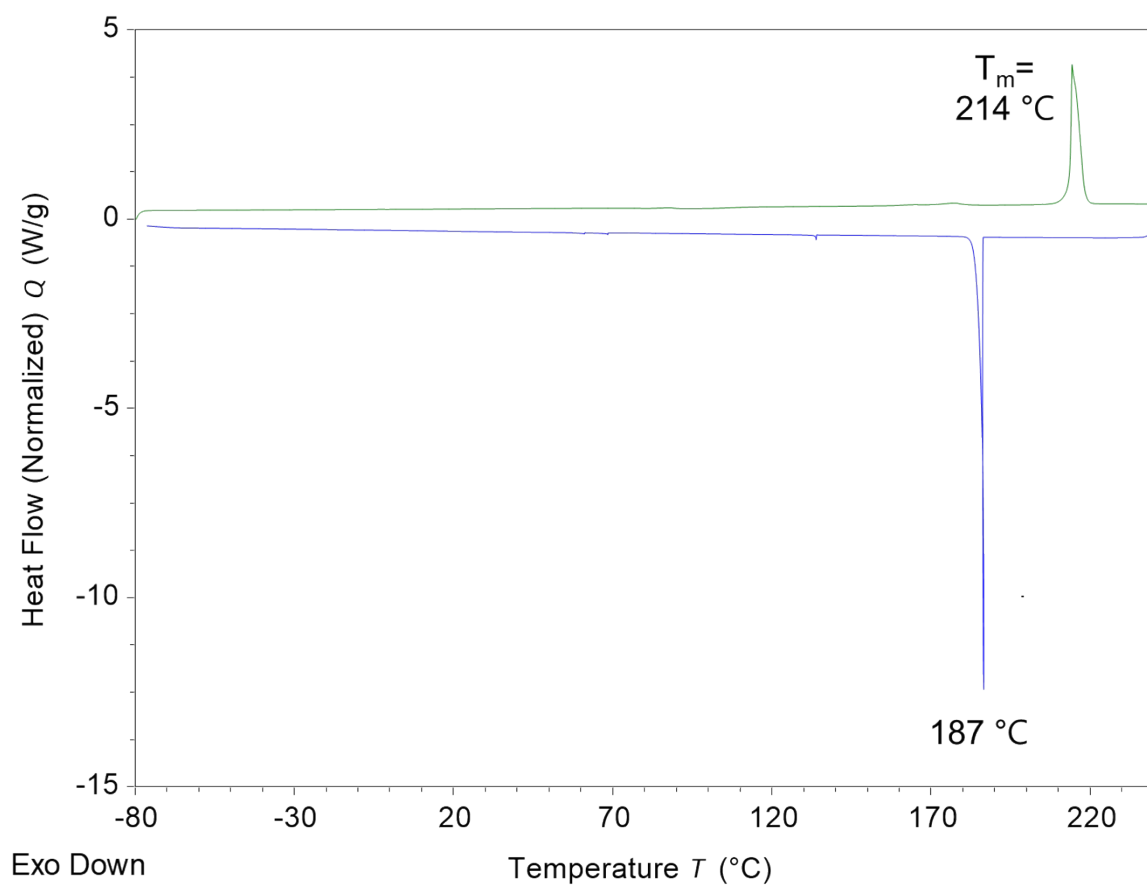


Figure S27. DSC 2nd heating (green) and cooling (blue) curves of **2-PF₆**

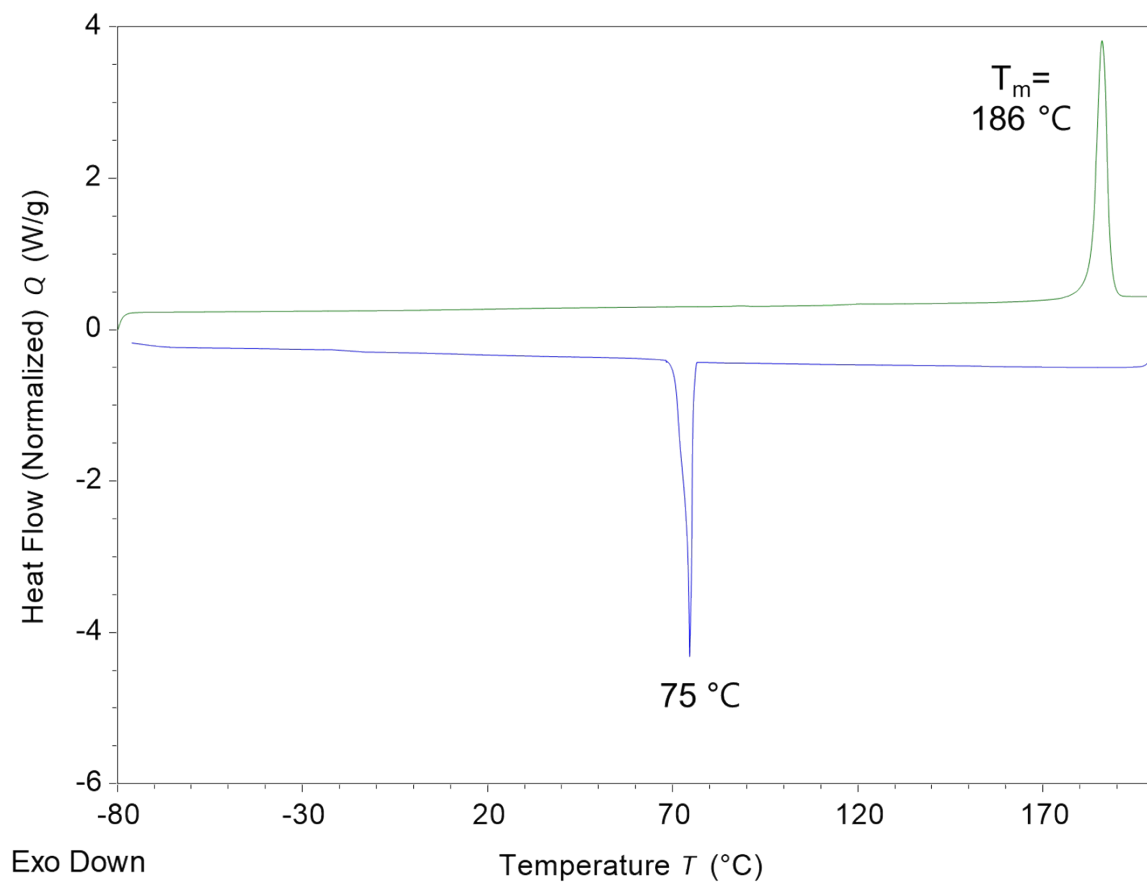


Figure S28. DSC 2nd heating (green) and cooling (blue) curves of **3-PF₆**

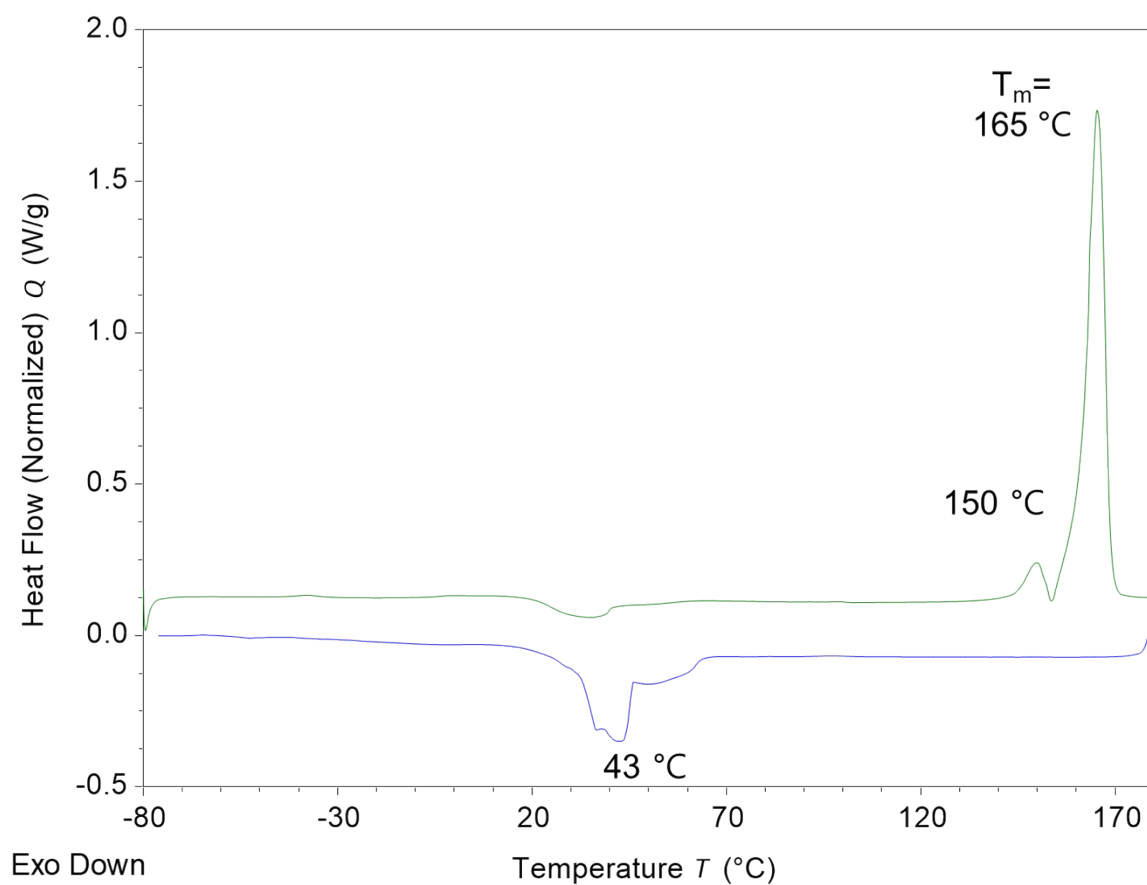


Figure S29. DSC 2nd heating (green) and cooling (blue) curves of **4-PF₆**

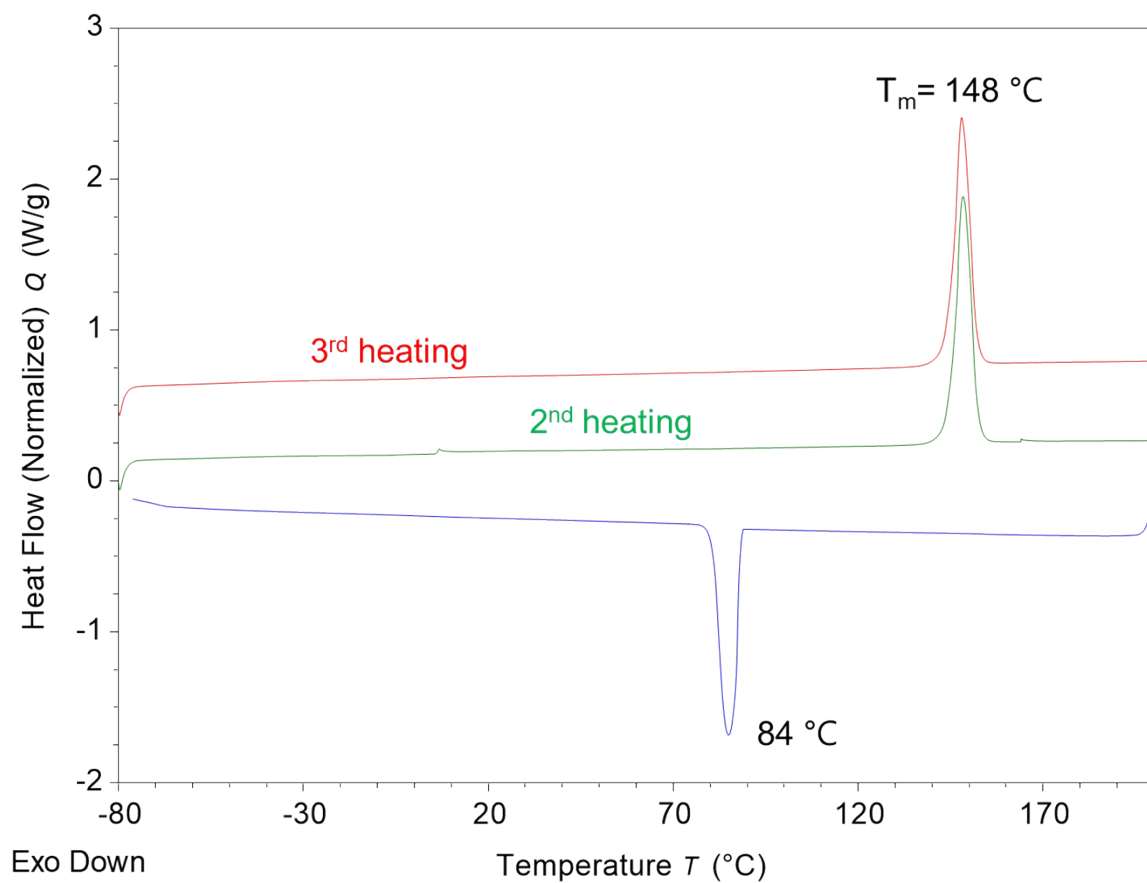


Figure S30. DSC 3rd heating (red), 2nd heating (green) and cooling (blue) curves of **5-PF₆**

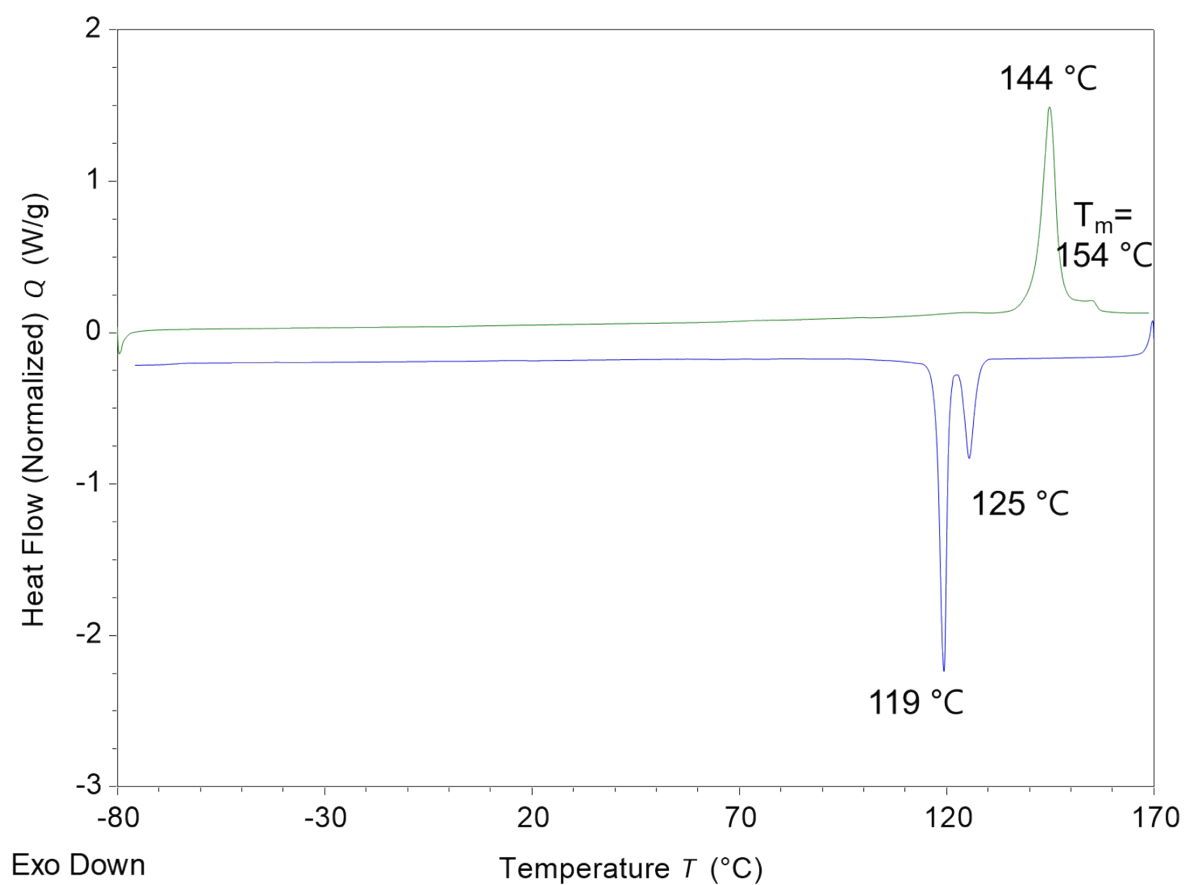


Figure S31. DSC 2nd heating (green) and cooling (blue) curves of **6-PF₆**

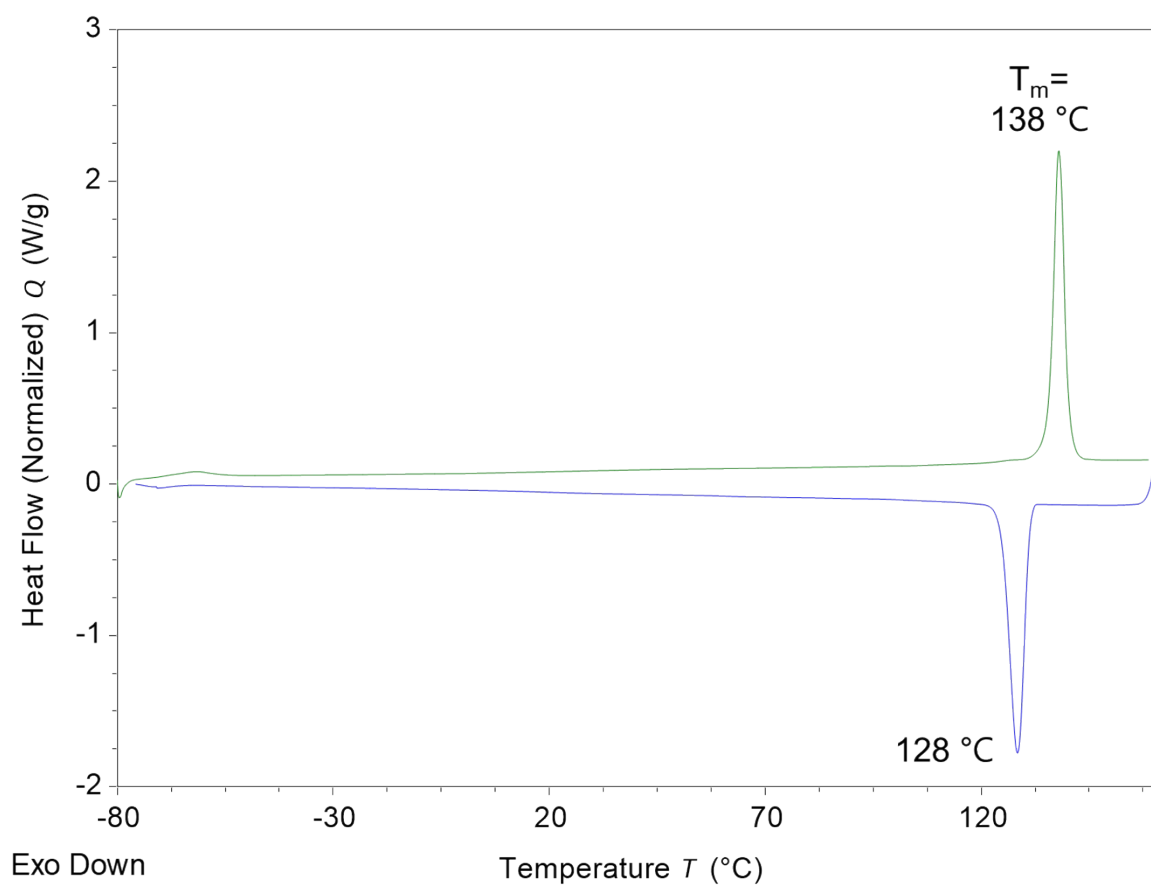


Figure S32. DSC 2nd heating (green) and cooling (blue) curves of **7-PF₆**

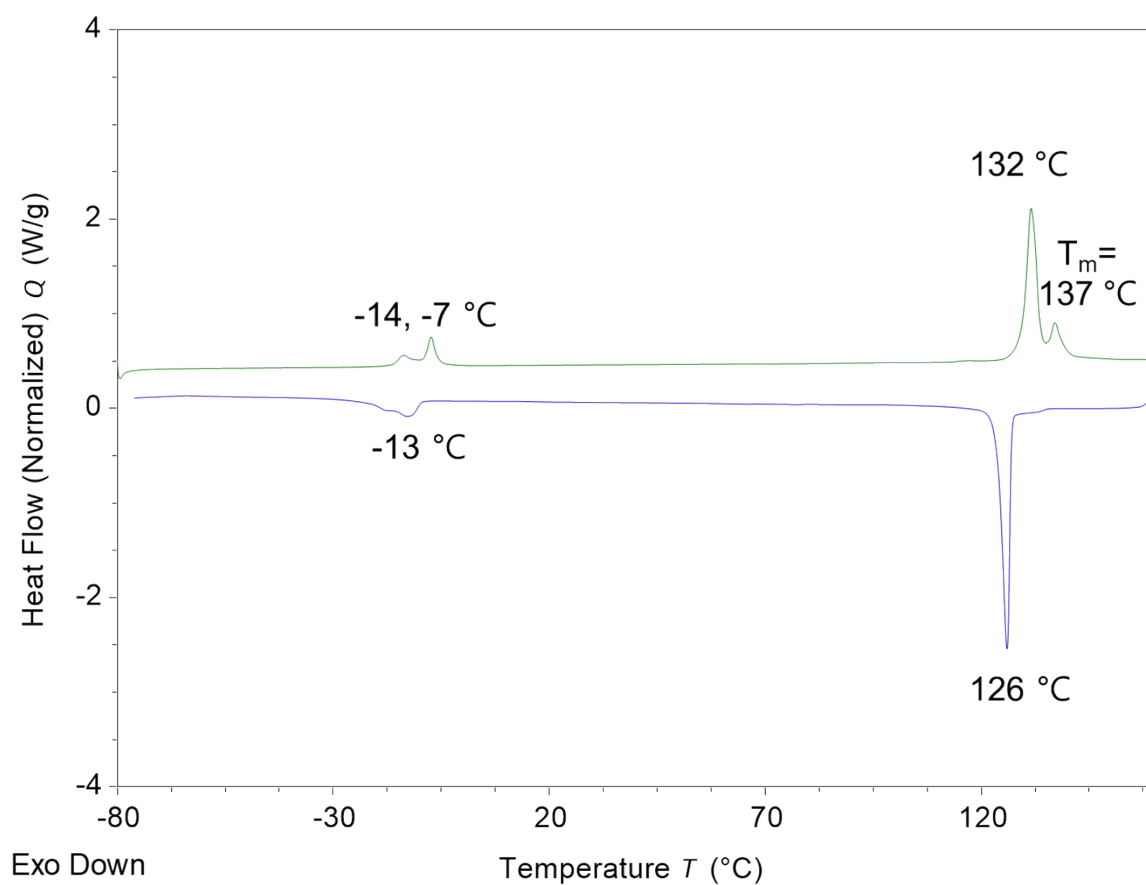


Figure S33. DSC 2nd heating (green) and cooling (blue) curves of **8-PF₆**

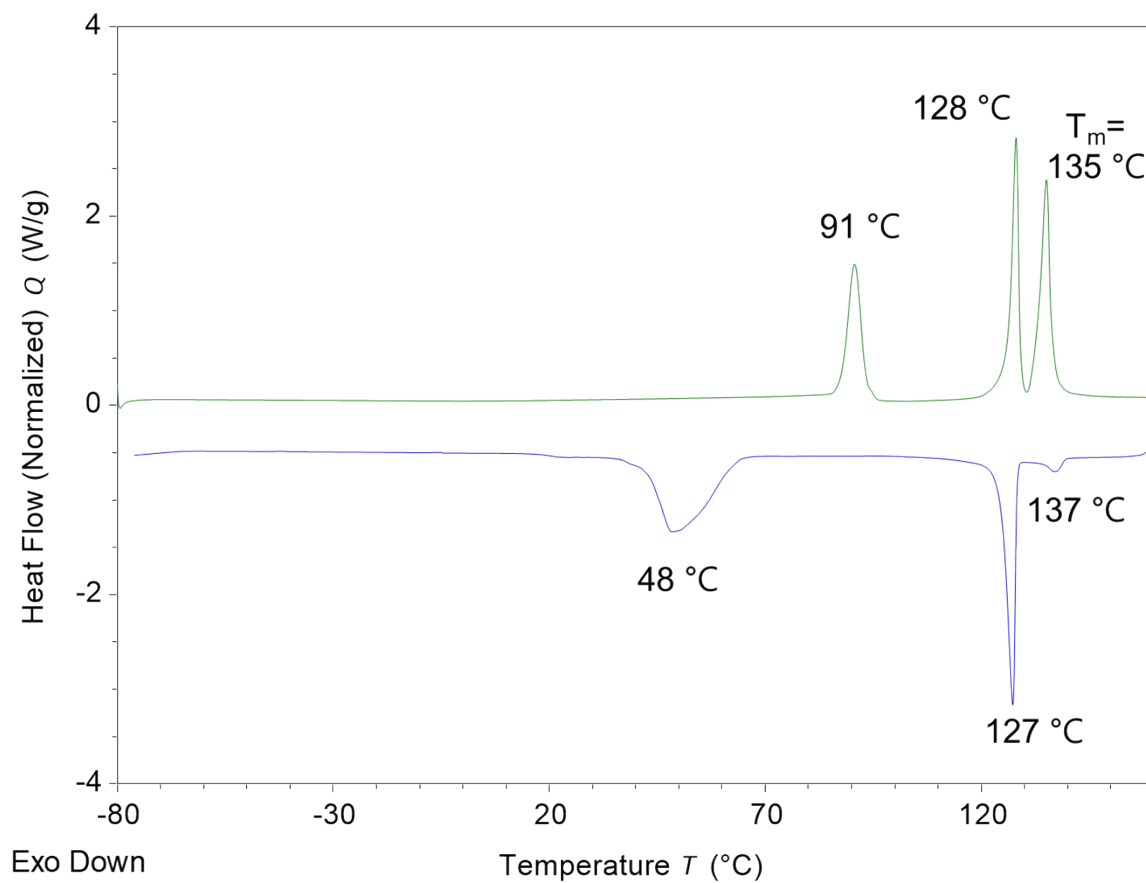


Figure S34. DSC 2nd heating (green) and cooling (blue) curves of **9-PF₆**

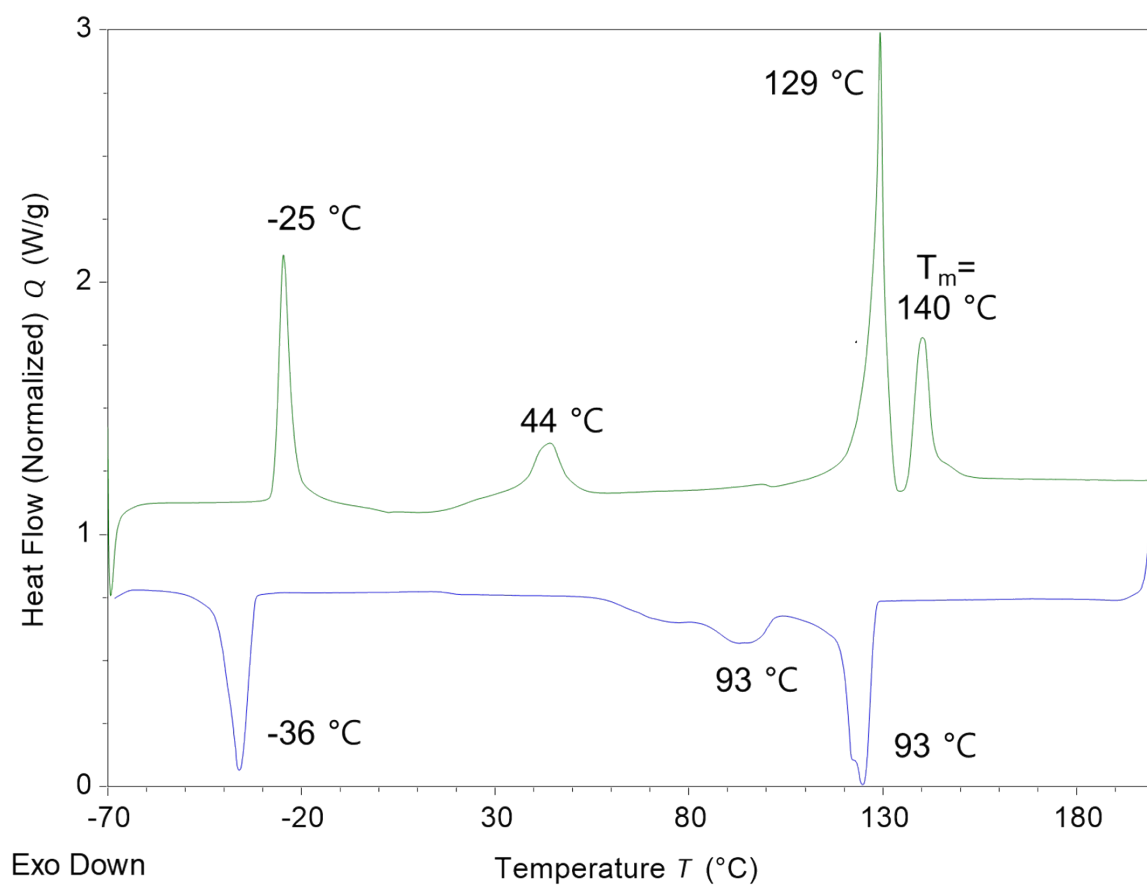


Figure S35. DSC 2nd heating (green) and cooling (blue) curves of **10-PF₆**

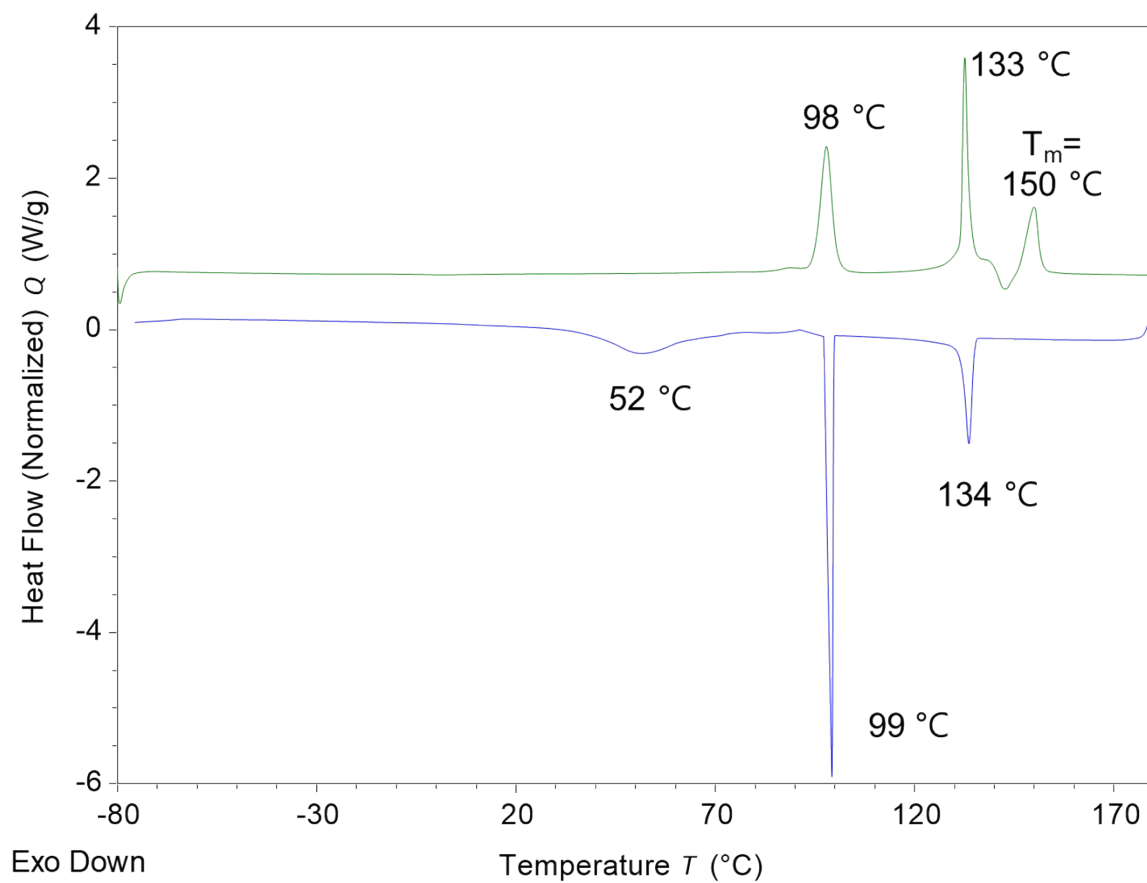


Figure S36. DSC 2nd heating (green) and cooling (blue) curves of **11-PF₆**

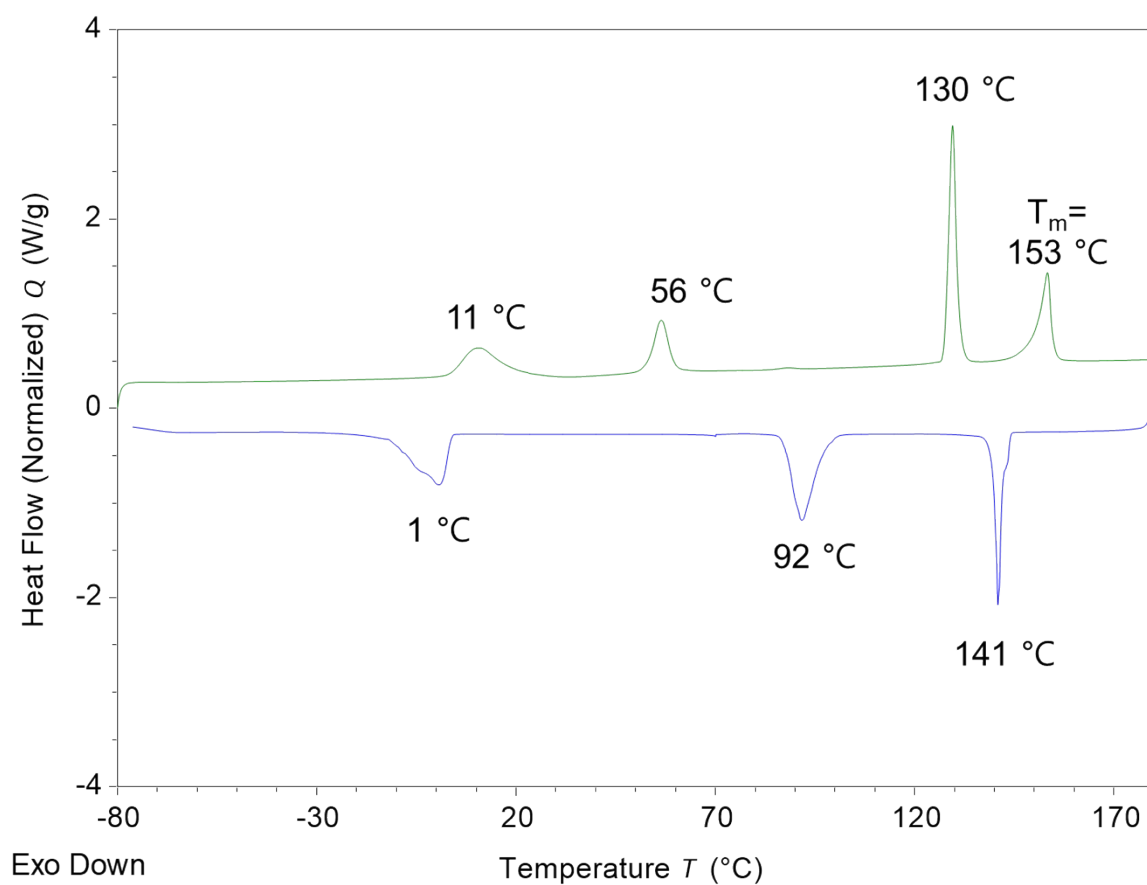


Figure S37. DSC 2nd heating (green) and cooling (blue) curves of **12-PF₆**

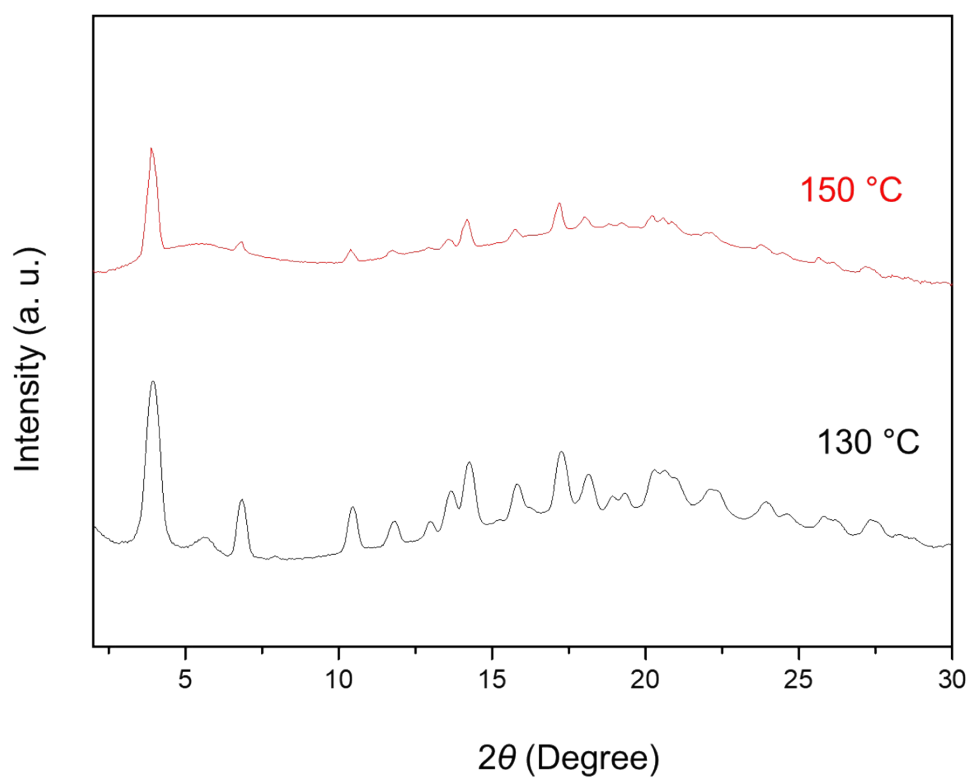


Figure S38. 1D wide-angle X-ray scattering spectra of (a) **6-PF₆**

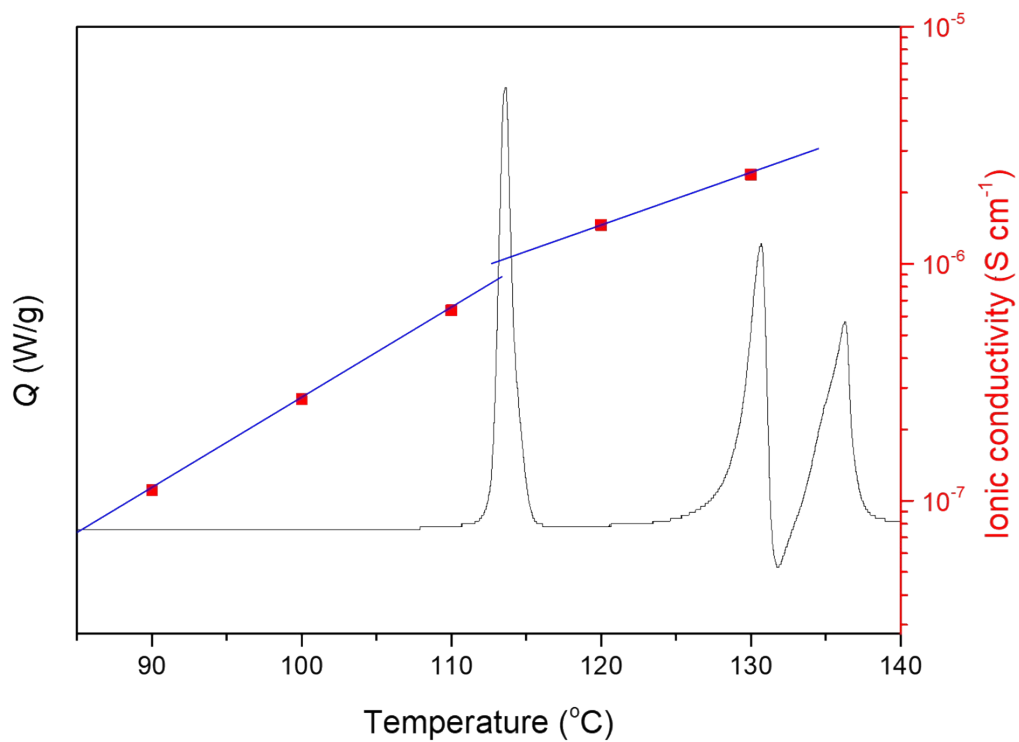


Figure S39. Temperature dependence of ionic conductivity and DSC trace of heating scan (rate = 1 °C·min⁻¹; N₂) of 8-PF₆

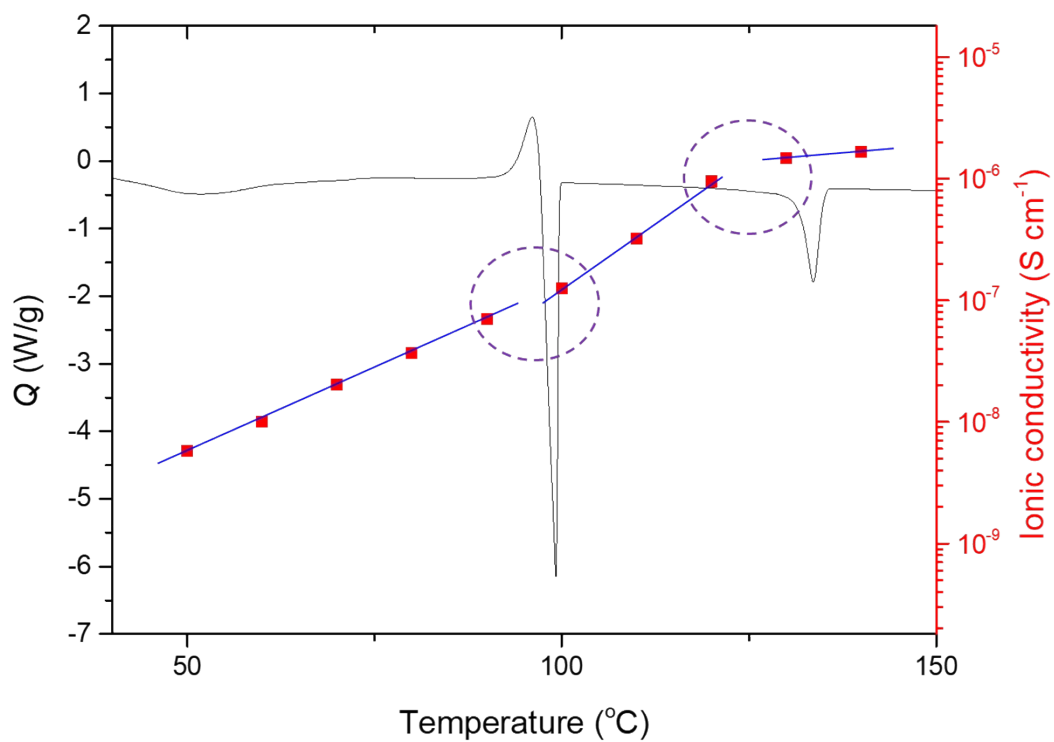


Figure S40. Temperature dependence of ionic conductivity and DSC trace of cooling scan (rate = 10 °C·min⁻¹; N₂) of 11-PF₆

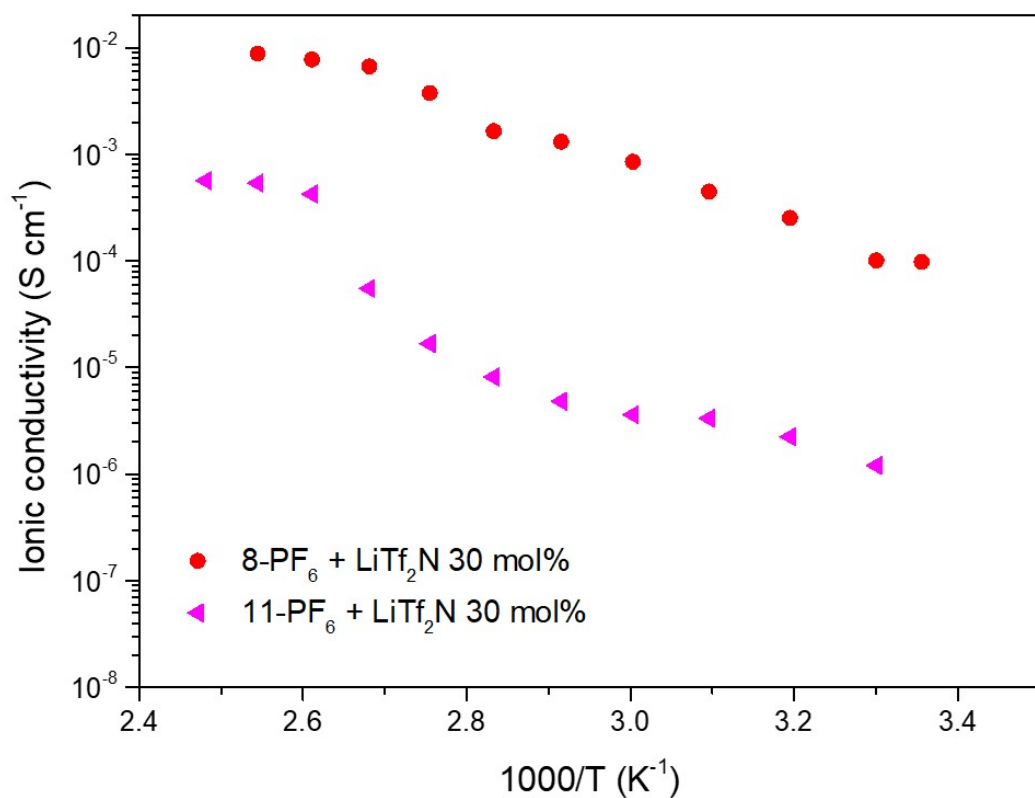


Figure S41. Temperature-dependent ionic conductivities of **8-PF₆** and **11-PF₆** solid electrolytes containing 30 mol% LiTf₂N measured by electrochemical impedance spectroscopy.

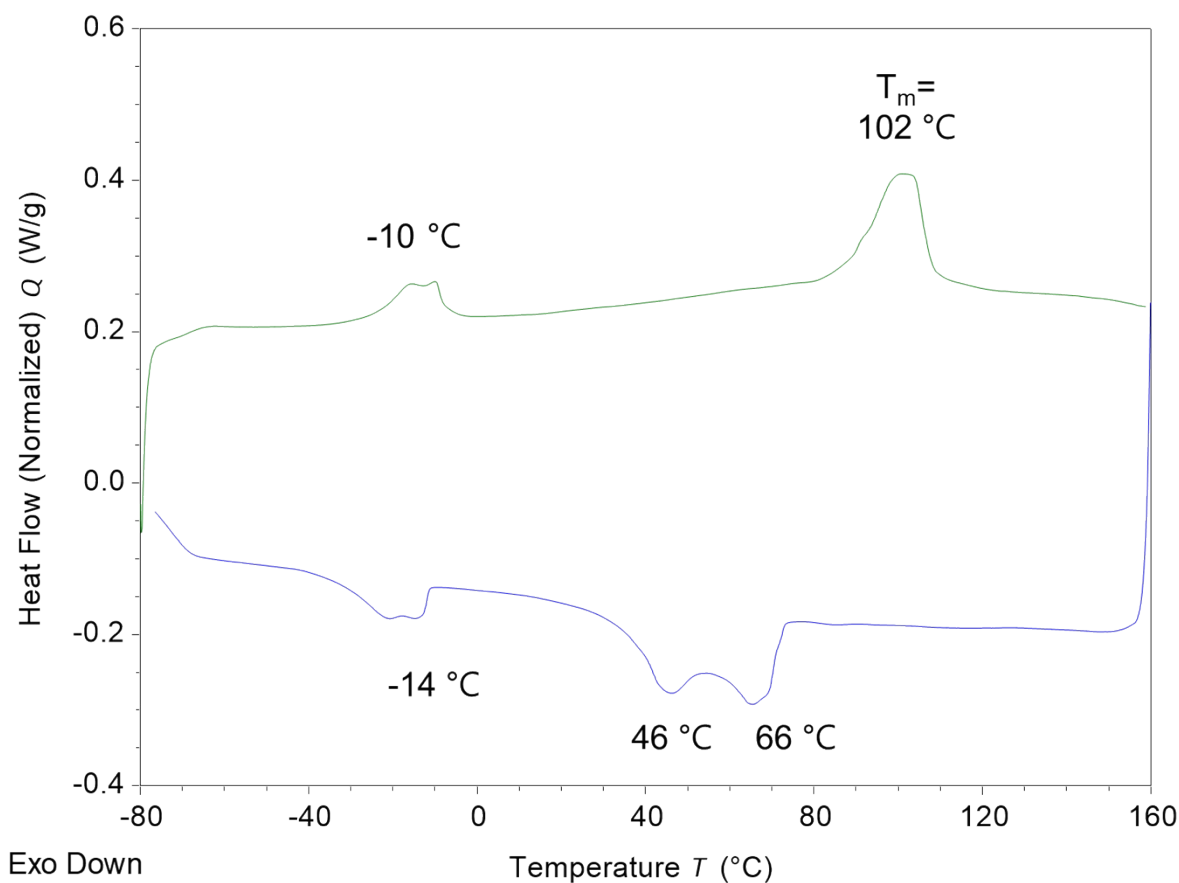


Figure S42. DSC 2nd heating (green) and cooling (blue) curves of **8-PF₆** + LiTf₂N 30 mol%

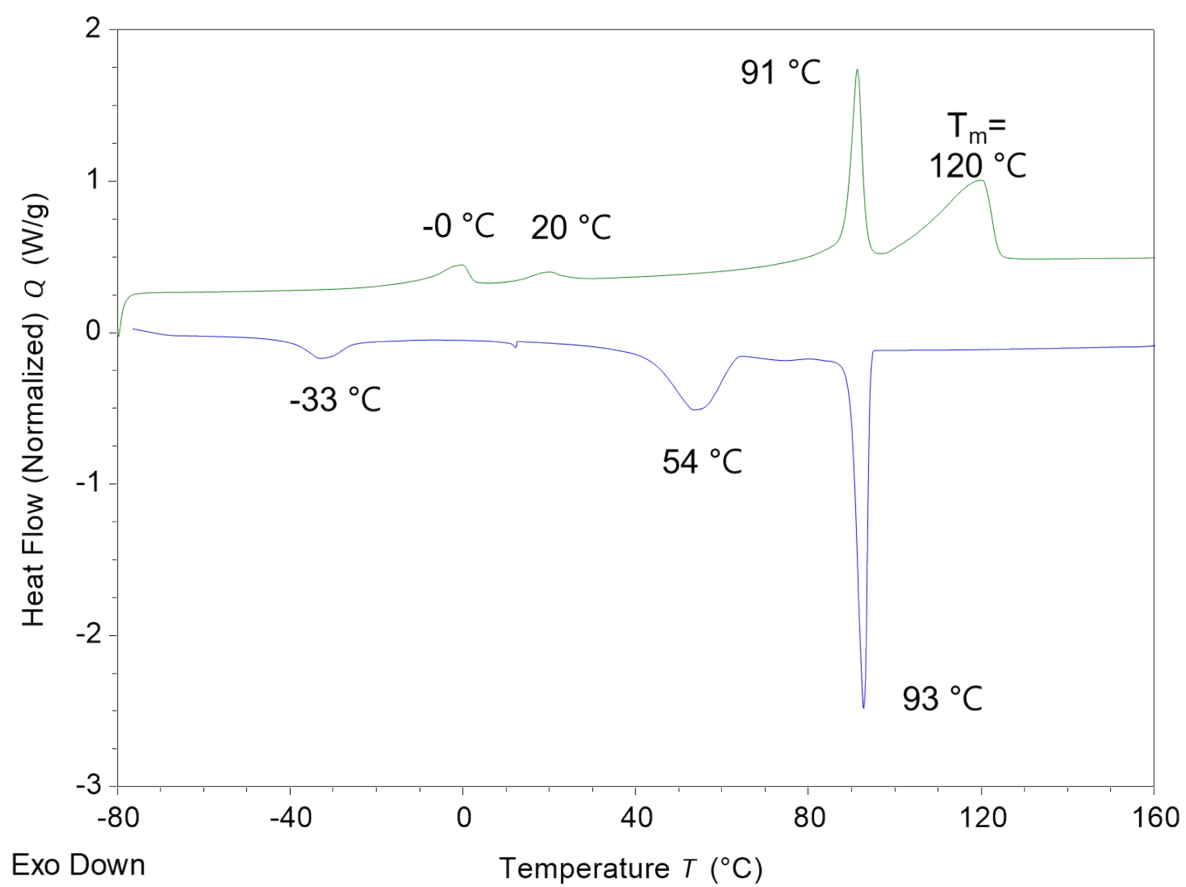


Figure S43. DSC 2nd heating (green) and cooling (blue) curves of **11-PF₆ + LiTf₂N 30 mol%**