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Supplementary Information

Plastic Crystal-based Electrolytes Using Novel Dicationic Salts

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Figure S 1. Arrhenius plot of conductivity for neat [C₂-Pyrr2][TFSI]₂ and neat [C₂-Pyrr3][TFSI]₂.



Figure S 2. ¹H static NMR of neat [C₂-Pyrr2][TFSI]₂



Figure S 3. ¹⁹F static NMR of neat [C₂-Pyrr2][TFSI]₂



Figure S 4. 1 H static NMR of neat [C₂-Pyrr3][TFSI]₂



Figure S 5. ¹⁹F static NMR of neat [C₂-Pyrr3][TFSI]₂



Figure S 6. ¹H NMR relative intensity of neat $[C_2$ -Pyrr2][TFSI]_2 recorded at increasing temperatures. The phase II to II transition is marked with a shaded area.



Figure S 7. ¹H NMR relative intensity of neat $[C_2$ -Pyrr2][TFSI]_2 and of neat $[C_2$ -Pyrr3][TFSI]_2 recorded at increasing temperatures. The III to II phase transition of pure $[C_2$ -Pyrr3][TFSI]_2 is shown by the clear region and the II to I phase transition of each system marked by the shaded region.



Figure S 8. ¹H static NMR of [C₂-Pyrr2][TFSI]₂/LiTFSI (10 mol % LiTFSI).



Figure S 9. ¹⁹F static NMR of [C₂-Pyrr2][TFSI]₂/LiTFSI (10 mol % LiTFSI).



Figure S 10. 7 Li static NMR of [C₂-Pyrr2][TFSI]₂/LiTFSI (10 mol % LiTFSI).



Figure S 11. ¹H static NMR of [C₂-Pyrr3][TFSI]₂/LiTFSI (10 mol % LiTFSI).



Figure S 12. ¹⁹F static NMR of [C₂-Pyrr3][TFSI]₂/LiTFSI (10 mol % LiTFSI).



Figure S 13. ⁷Li static NMR of $[C_2$ -Pyrr3][TFSI]₂/LiTFSI (10 mol % LiTFSI).



Figure S 14. (a) and (b) $^1\!\mathrm{H}$ static NMR fitting using DMFIT software.



Figure S 15. (a) and (b) $^{19}\mathrm{F}$ static NMR fitting using DMFIT software.



Figure S 16. (a) and (b) ⁷Li static NMR fitting using DMFIT software.



Figure S 17. ¹H NMR line widths of pure $[C_2$ -Pyrr2][TFSI]_2 and $[C_2$ -Pyrr2][TFSI]_2/LiTFSI (10 mol % LiTFSI) mixture recorded at increasing temperatures. The phase transition of both pure $[C_2$ -Pyrr2][TFSI]_2 (80–100 °C) and $[C_2$ -Pyrr2][TFSI]_2/LiTFSI is marked with a shaded area.



Figure S 18. ¹H NMR relative intensity of pure $[C_2$ -Pyrr2][TFSI]_2 and $[C_2$ -Pyrr2][TFSI]_2/LiTFSI (10 mol% LiTFSI) mixture recorded at increasing temperatures. The phase transition of both pure $[C_2$ -Pyrr2][TFSI]_2 (80–100 °C) and $[C_2$ -Pyrr2][TFSI]_2/LiTFSI is marked with a shaded area.



Figure S 19. ¹H NMR line widths of $[C_2$ -Pyrr3][TFSI]_2/LiTFSI (10 mol % LiTFSI) recorded at increasing temperatures. The phase transition for both pure $[C_2$ -Pyrr3][TFSI]_2 and $[C_2$ -Pyrr3][TFSI]_2/LiTFSI is shown as S*: 65–80 °C and S**: 80–100 °C.



Figure S 20. ¹H NMR relative intensity of $[C_2$ -Pyrr3][TFSI]₂/LiTFSI (10 mol % LiTFSI) recorded at increasing temperatures. The phase transition for both pure $[C_2$ -Pyrr3][TFSI]₂ and $[C_2$ -Pyrr3][TFSI]₂/LiTFSI is shown as S*: 65–80 °C and S**: 80–100 °C.



Figure S 21. (a) ¹⁹F NMR line width of pure $[C_2$ -Pyrr3][TFSI]_2 and $[C_2$ -Pyrr3][TFSI]_2/LiTFSI (10 mol % LiTFSI) and (b) ¹⁹F NMR relative intensity of $[C_2$ -Pyrr3][TFSI]_2/LiTFSI (10 mol % LiTFSI) recorded at increasing temperatures. The phase transition for both pure pure $[C_2$ -Pyrr3][TFSI]_2 and $[C_2$ -Pyrr3][TFSI]_2/LiTFSI is shown as S+: 65–80 °C and S++: 80–100 °C. For FWHM and relative intensity analysis, CSA model and a Gaussian model (for the narrow component) were used in combination for peak fitting.



Figure S 22. ⁷LiNMR line widths of $[C_2$ -Pyrr2][TFSI]_2/LiTFSI and $[C_2$ -Pyrr3][TFSI]_2/LiTFSI recorded at increasing temperatures. The 68–80 °C phase transition of $[C_2$ -Pyrr3][TFSI]_2/LiTFSI is shown by the clear region and the 80–100 °C phase transition of each system marked as the shaded region.