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Development of organic ionic plastic crystals composed of tetrahydrothiophenium cation with high conductivity

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Ionic plastic crystals (IPCs) are promising candidates for solid electrolytes due to their plasticity and ionic conductivity. While pyrrolidinium-based IPCs from five-membered heterocycles have been synthesized, their conductivities remain below practical levels. Here, tetrahydrothiophene was selected as a novel structural candidate due to its similarity to pyrrolidinium, and new IPCs were synthesized and evaluated for their physicochemical properties.

In this study, organic salts were synthesized as precursors by reacting tetrahydrothiophene with alkyl iodides. Subsequent anion exchange reactions between these precursors and alkali metal salts in acetone yielded [S*n*][CF_3BF_3] (*n* = 1–3) (Fig. 1). To investigate their physicochemical properties, differential scanning calorimetry (DSC) and AC impedance measurements were conducted to determine their phase transition temperatures and ionic conductivities.

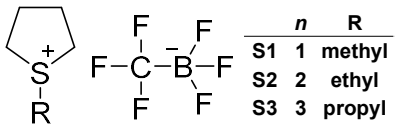

Fig 1. Chemical structure of [S*n*][CF_3BF_3] (*n* = 1–3).

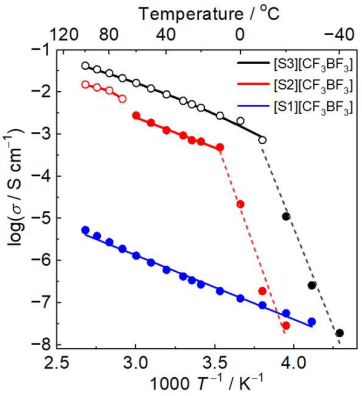

Fig 2. Arrhenius plots of ionic conductivities for [S*n*][CF_3BF_3] (*n* = 1–3). Open plots: liquid, Closed plots: solid.

DSC measurements showed that [S1][CF_3BF_3] had a solid–solid transition at −24.1 °C, with no detectable melting point. [S2][CF_3BF_3] exhibited transitions at −73.2, −17.3, and 12.6 °C, and melted at 77.4 °C. Its entropy of fusion was 0.31 J K^{-1} mol^{-1}, below Timmermans' criterion of 20 J K^{-1} mol^{-1}, suggesting that [S2][CF_3BF_3] is an IPC. [S3][CF_3BF_3] melted at −4.9 °C and remained liquid at room temperature, indicating that it is an ionic liquid.

Fig. 2 shows the Arrhenius plots of ionic conductivities for [S*n*][CF_3BF_3] (*n* = 1–3). AC impedance measurements revealed that [S2][CF_3BF_3] exhibited a high ionic conductivity of 7.19 × 10^{-4} S cm^{-1} at 25 °C, which is about ten times higher than that of conventional pyrrolidinium-based IPCs.

References

1. Yamada, H.; Miyachi, Y.; Takeoka, Y.; Rikukawa, M.; Yoshizawa-Fujita, M., *Electrochim. Acta.* 2019, *303*. 293

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