

## RESEARCH ARTICLE OPEN ACCESS

# Eutectic Engineering of Ionic-Liquid Electrolytes for Enhanced Organic Battery Performance

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**Received:** 5 August 2026 | **Revised:** 5 September 2026 | **Accepted:** 23 September 2026

**Keywords:** deep eutectic solvents | electrochemistry | hydrogen-bond donor | ionic liquids | organic battery | polymers | redox-active polymers | sustainable chemistry

## ABSTRACT

Ionic liquids are attractive electrolytes for organic batteries because of their excellent electrochemical and thermal stability, yet their high viscosity severely limits ionic transport and rate capability. Existing approaches mainly rely on dilution or additives but do not provide a general strategy for simultaneously improving conductivity while preserving electrolyte stability. Here, we demonstrate that the design principles of deep eutectic solvents can be transferred to ionic-liquid electrolytes to rationally tailor their physicochemical and electrochemical properties. By introducing the hydrogen-bond donor *N*-methylacetamide (NMA) into the ionic liquid 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([EMIm][TFSI]), a eutectic composition was identified through differential scanning calorimetry. After optimization, the electrolyte exhibits a 45% reduction in viscosity at room temperature while maintaining a wide electrochemical stability window of 4.18 V and increasing the ionic conductivity at 25 °C from 0.38 to 0.88 S · m<sup>-1</sup>. When employed in PTMA polymer-based half cells, the eutectic electrolyte enables faster electrochemical activation, enhanced specific currents, improved capacity retention, and stable operation over 500 cycles. These findings establish deep eutectic interactions as an effective molecular engineering strategy for tailoring ionic-liquid electrolytes and provide a versatile platform for developing safer and higher-performance electrolytes for next-generation organic batteries.

## 1 | Introduction

One of the major challenges in the transition toward sustainable energy technologies is the development of electrochemical energy storage systems that combine high performance with long-term environmental sustainability. Although lithium-ion batteries have revolutionized portable electronics and electric transportation, their continued expansion raises concerns regarding the availability of critical raw materials [1–3], the environmental footprint associated with mining and processing transition metals [1, 3, 4], and the complexity of recycling multi-component inorganic electrodes [2, 5]. Consequently, increasing attention has been directed toward alternative battery chemistries that rely on

earth-abundant resources and more sustainable materials while maintaining competitive electrochemical performance [6].

Among the various emerging technologies, polymer-based organic batteries represent a particularly attractive platform because both the active materials and, potentially, the electrolyte can be synthesized from abundant organic building blocks [1, 7, 8]. Redox-active polymers offer several advantages, including structural versatility [9], lightweight construction [10], mechanical flexibility [11, 12], and the possibility of tailoring their electrochemical properties through molecular design [13]. These characteristics make them promising candidates for wearable electronics [14, 15], flexible devices [16], and

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sustainable stationary energy storage [3, 16]. Significant progress has been achieved in the development of highly stable radical polymers such as poly(2,2,6,6-tetramethylpiperidinyloxy methacrylate) (PTMA), which exhibits rapid redox kinetics, excellent reversibility, and high theoretical capacities [16–18]. Despite these advances, the practical performance of polymer batteries remains largely dictated by the electrolyte, which controls ionic transport, electrode wetting, electrochemical stability, and the long-term preservation of the active material [10, 19].

Electrolytes developed for conventional lithium-ion batteries are often unsuitable for organic electrode materials. Carbonate- and ether-based electrolytes exhibit high volatility and flammability, creating significant safety concerns under abusive operating conditions [20, 21]. Moreover, many organic redox-active compounds display partial solubility in these solvents, resulting in continuous loss of active material, self-discharge, and poor cycling stability [10, 20, 22]. These limitations become particularly severe for polymer electrodes, where electrolyte-induced degradation frequently determines the achievable lifetime of the battery. Consequently, identifying alternative electrolyte concepts has become one of the central challenges in the development of high-performance organic batteries [10].

Ionic liquids have emerged as one of the most promising alternatives because of their negligible vapor pressure, nonflammability, high thermal stability, and broad electrochemical stability windows [8, 10, 20, 23, 24]. Numerous studies have demonstrated that ionic liquids suppress polymer dissolution [25, 26], improve cycling stability [27, 28], and enable safer battery operation compared with conventional molecular solvents [10]. Nevertheless, their practical implementation remains hindered by their intrinsically high viscosity [5, 23, 24]. Strong electrostatic interactions between the constituent ions reduce molecular mobility, resulting in sluggish ion transport, slow electrode wetting, elevated interfacial resistance, and poor rate capability, particularly at room temperature [29]. Although elevated operating temperatures [30–32], cosolvents, and electrolyte additives have been investigated to mitigate these limitations [33], these approaches often compromise either safety or electrochemical stability and do not provide a general framework for rational electrolyte design.

An alternative strategy may arise from the concepts underlying deep eutectic solvents (DESs). DESs are formed through strong hydrogen-bond interactions between hydrogen-bond donors and acceptors, producing liquid phases whose physicochemical properties differ substantially from those of the individual components. Beyond serving as electrolytes themselves, DES chemistry demonstrates how intermolecular interactions can be deliberately engineered to tailor viscosity, conductivity, thermal behavior, and electrochemical properties [5, 34–37]. Despite the rapid development of DES electrolytes for various electrochemical applications, these molecular design principles have rarely been exploited to engineer conventional ionic-liquid electrolytes. This represents a significant opportunity because introducing controlled hydrogen-bond interactions into ionic liquids could weaken ion–ion interactions, enhance molecular mobility, and improve ionic transport while preserving the intrinsic advantages of ionic-liquid systems.

Here, we hypothesize that the concepts governing DES formation can be transferred to ionic-liquid electrolytes to rationally tune their transport properties without sacrificing electrochemical stability. Specifically, we propose that introducing an appropriate hydrogen-bond donor into an ionic liquid can partially disrupt the ionic network responsible for its high viscosity, thereby improving ion mobility while maintaining the wide electrochemical stability window required for polymer batteries. Rather than considering DESs as replacement electrolytes, this approach uses DES chemistry as a molecular engineering strategy for optimizing existing ionic-liquid electrolytes. To test this concept, we investigate binary mixtures of 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([EMIm][TFSI]) and N-methylacetamide (NMA). The eutectic composition is identified through thermal analysis and correlated with changes in viscosity, ionic conductivity, and electrochemical stability. The optimized electrolyte is subsequently evaluated in PTMA-based half-cells to establish relationships between electrolyte structure, transport properties, and electrochemical performance. We demonstrate that the eutectic composition provides the optimum balance between ionic conductivity, viscosity, electrochemical stability, and long-term cycling behavior. In the broader context of low-melting mixtures, recently discussed under the term low-melting multi-component mixtures (LoMMs), this study demonstrates how eutectic interactions can be exploited to tailor an ionic-liquid electrolyte while retaining its essential characteristics [38].

## 2 | Experimental Procedures

### 2.1 | Electrolyte Preparation

[EMIm][TFSI] (99.9%) (proionic) was obtained from Carl Roth and NMA (99.9%) was procured by Sigma–Aldrich. The NMA was distilled under vacuum and then dried with 3 and 4 Å molecular sieves for over 2 weeks. The water content was recorded using Karl Fischer titration and came out to be 7 ppm. This NMA was heated at 70 °C using a heating plate until it was completely liquid. Different compositions were then made using different masses of [EMIm][TFSI] and NMA corresponding to precalculated molar ratios of the two components. The calculated amounts of [EMIm][TFSI] and NMA were added to vials, which were heated at 80 °C for 2 h afterwards and kept on stirring overnight. Molecular sieves were added to each composition to dry them for at least 1 week. The mixtures were visibly liquid and were ready for further measurements. The entire preparation, including the drying, was done inside a nitrogen-flooded glovebox.

### 2.2 | Electrode Preparation

PTMA particles (average particle size: 100 nm, degree of oxidation: 77 ± 12%) were provided by Friedrich Schiller University Jena. The electrode slurry was prepared by dispersing PTMA, Super P conductive carbon (Alfa Aesar), and sodium carboxymethyl cellulose (CMC, Sigma–Aldrich) in deionized water (conductivity: 0.055 μS · cm<sup>−1</sup>) at a mass ratio of 35:60:5 using an Ultra-Turrax T8 disperser (IKA). In other words, the electrode slurry consisted of 35 wt% PTMA, 60 wt% Super P conductive carbon, and 5 wt% CMC binder. The resulting slurry was allowed

to equilibrate for at least 30 min before being homogenized again at maximum speed for 2–10 min, depending on the batch volume. Prior to coating, aluminum foil (Korff, 15  $\mu\text{m}$  thickness) was pretreated in an aqueous KOH solution (pH 12) for 1 min, rinsed if applicable, and dried. The homogeneous slurry was then doctor-bladed onto the pretreated aluminum foil and allowed to level at room temperature for approximately 5 min before drying at 80 °C in a drying oven. Circular electrodes with a diameter of 12 mm were punched from the dried electrode film and used for all electrochemical measurements.

## 2.3 | Thermal Measurements

Thermal measurements were performed with a differential scanning calorimeter (DSC) from TA Instruments (T250). Approximately 4  $\mu\text{L}$  of the mixture was added into preweighed Al pans and covered with hermetic lids, which were sealed using the TA sample press to form DSC samples for each composition. These pans were weighed again to obtain the sample weight. All compositions were then put under the same procedure in the DSC. The samples were heated to 80 °C at a rate of 2 °C/min, kept at 80 °C for 5 min to ensure complete erasure of previous thermal history, and then cooled to –80 °C at the rate of 2 °C  $\cdot$  min<sup>–1</sup>. After keeping the samples at –80 °C for 5 min, they were again heated to 80 °C at 2 °C  $\cdot$  min<sup>–1</sup>. The last step of heating was then used to show the melting curves and, in some cases, the cold crystallization. These thermal graphs were used to formulate the phase diagram and the Tammann plot for this binary system. The samples were also weighed after the measurement to ensure that no mixture had leaked during the measurement.

## 2.4 | Viscosity Measurements

Viscosity measurements were performed using an Anton Paar SVM 3001 smart viscometer. Around 2 mL of the mixture was degassed and then slowly injected into the viscometer tube, ensuring the absence of bubbles. The density, kinematic viscosity, and dynamic viscosity were determined over a temperature range of 20 °C–80 °C (i.e., 20, 25, 30, 40, 50, 60, 70, and 80 °C) under equilibrium conditions according to the instrument's standard measurement protocol.

## 2.5 | Conductivity Measurements

The conductivity values were determined through electrochemical impedance spectroscopy (EIS) using Bio-Logic MPG2 Potentiostat. Three-electrode Swagelok-type cell was used for these measurements with PTMA polymer electrode as working electrode, a graphite counter electrode, and an annealed silver wire as pseudo-reference electrode. The cell was kept at open circuit potential (OCP) for 2 h in BINDER FP 53 climate chamber at 25 °C, after which the EIS was measured. The temperature was then further increased to 35, 45, 55, 65, and 75 °C, and an EIS measurement was recorded at each step. The cell was kept at each mentioned temperature for 1 h to stabilize. This measurement was performed for [EMIm][TFSI] and the two compositions from [EMIm][TFSI]:NMA mixture, which were the main focus of this study.

## 2.6 | Electrochemical Measurements

The electrochemical measurements were performed using a Gamry Interface 1010B Potentiostat. Linear sweep voltammetry was performed with a Swagelok-type three-electrode setup with a glassy carbon working electrode, a graphite counter electrode, and an annealed silver wire as pseudo-reference electrode. The cyclic voltammetry and galvanostatic cycling with the mixtures were done using similar setups as mentioned above, with the PTMA polymer electrode as the working electrode. All cells were kept at OCP for 2 h before the cyclic voltammetry and the galvanostatic cycling measurements. The cell assembly and the measurements were performed inside a nitrogen-flooded glovebox.

## 3 | Results and Discussion

### 3.1 | Engineering the Eutectic Electrolyte

To explore whether DES chemistry can be employed as a molecular design strategy for tailoring ionic-liquid electrolytes, binary mixtures of [EMIm][TFSI] and NMA are systematically investigated by differential scanning calorimetry (DSC). The objective is to identify the eutectic composition and establish the relationship between the thermal behavior and the subsequent electrochemical properties.

Figure 1a shows the DSC thermograms of the investigated mixtures. For NMA-rich compositions ( $0.033 \leq \chi_{[\text{EMIm}][\text{TFSI}]} \leq 0.476$ ), the melting transition of pure [EMIm][TFSI] remains visible at approximately –17 °C, while the melting temperature of the NMA-rich phase continuously increases with increasing NMA content. In contrast, mixtures containing higher ionic-liquid fractions ( $0.500 \leq \chi_{[\text{EMIm}][\text{TFSI}]} \leq 0.935$ ) exhibit significantly more complex thermal behavior characterized by multiple melting events (Figure S1) and cold-crystallization processes (Figures S2–S4). The overlapping melting transitions indicate that the [EMIm][TFSI]/NMA system deviates considerably from ideal mixing and undergoes composition-dependent structural rearrangements.

The phase diagram constructed from the DSC data (Figure 1b) further demonstrates the nonideal character of the binary system. Several composition-dependent phase regions are observed, reflecting the coexistence of different crystalline and liquid phases. Compared with the previously reported NaTFSI/NMA system [39], the considerably more complex phase behavior is most likely associated with the larger steric demand and conformational flexibility of the imidazolium cation, which introduces additional intermolecular interactions absent in inorganic salt-based eutectic systems [23].

The lowest and single melting transition is observed at  $\chi_{[\text{EMIm}][\text{TFSI}]} = 0.667$ , corresponding to a molar ratio of [EMIm][TFSI]:NMA = 1:0.5, indicating the formation of the eutectic composition. This assignment is independently confirmed by the Tammann analysis (Figure 1d), where the eutectic melting enthalpy reaches a maximum at the same composition. At the eutectic point, only the eutectic phase contributes to the melting process, producing the highest enthalpy of fusion and confirming that this composition represents the thermodynamically preferred liquid structure.

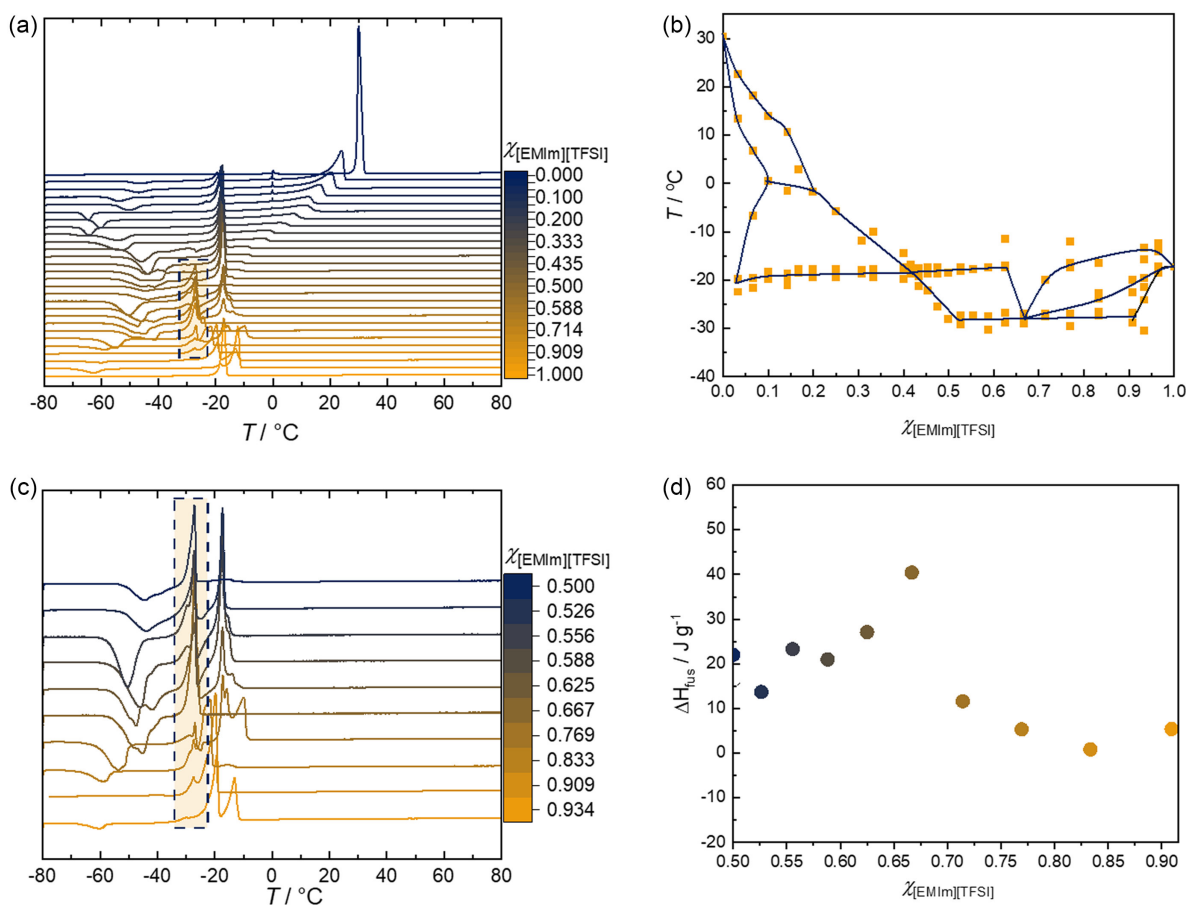


**FIGURE 1** | Thermal analysis showing (a) thermal graphs for different compositions, (b) phase diagram obtained from the thermal graphs (the lines are drawn as a guide for the eye), (c) zoomed area for the eutectic transition, and (d) Tammann plot obtained from eutectic transitions.

The eutectic behavior arises from a redistribution of intermolecular interactions upon mixing NMA with [EMIm][TFSI]. While the neat ionic liquid is dominated by strong electrostatic interactions between [EMIm]<sup>+</sup> and [TFSI]<sup>−</sup>, NMA introduces additional hydrogen-bonding and dipolar interactions that perturb this interconnected ionic network and promote nonideal mixing. This altered local interaction environment stabilizes the mixed liquid relative to the pure components, leading to the pronounced melting-point depression and formation of a distinct eutectic composition. Thus, the eutectic state is not merely a result of physical dilution but reflects a fundamental change in the molecular organization of the electrolyte. Identifying this composition is therefore important not only thermodynamically but also for optimizing transport properties as the balance between molecular mobility and structural organization directly influences viscosity, ionic conductivity, and electrochemical stability.

### 3.2 | Effect of Eutectic Interactions on Transport Properties

The high viscosity of ionic liquids remains one of the principal challenges limiting their application as battery electrolytes, as it restricts ion mobility, delays electrode wetting, and reduces rate capability [23, 29]. Having identified the eutectic composition, the next objective is therefore to determine whether the structural reorganization associated with eutectic formation effectively improves the transport properties of the electrolyte.

The temperature-dependent dynamic viscosities of neat [EMIm][TFSI], the eutectic composition ([EMIm][TFSI]:NMA = 1:0.5,  $\chi_{[EMIm][TFSI]} = 0.667$ ), and the equimolar mixture ([EMIm][TFSI]:NMA = 1:1,  $\chi_{[EMIm][TFSI]} = 0.500$ ) are shown in Figure 2a. For all compositions, viscosity decreases continuously with increasing temperature, as expected from the increased molecular mobility at elevated temperatures. More importantly, NMA substantially reduces the viscosity over the entire investigated temperature range. At 20 °C, the viscosity decreases from 40.05 mPa s for neat [EMIm][TFSI] to 22.20 mPa s for the eutectic composition, corresponding to a reduction of approximately 45%. Notably, the viscosity of the eutectic electrolyte at 20 °C is comparable to that of neat [EMIm][TFSI] at approximately 40 °C, demonstrating the pronounced effect of NMA incorporation on the liquid structure.

The reduction in viscosity is consistent with a change in the intermolecular interactions within the electrolyte. In neat [EMIm][TFSI], strong electrostatic interactions between the cation and anion contribute to an interconnected ionic environment that limits molecular rearrangement and flow. NMA introduces additional hydrogen-bonding and dipolar interactions that compete with these ionic interactions and modify the local liquid structure. This restructuring reduces the resistance to molecular motion and consequently facilitates flow. The viscosity results therefore provide macroscopic evidence that NMA substantially alters the interaction network of the ionic liquid rather than merely acting as an inert diluent.

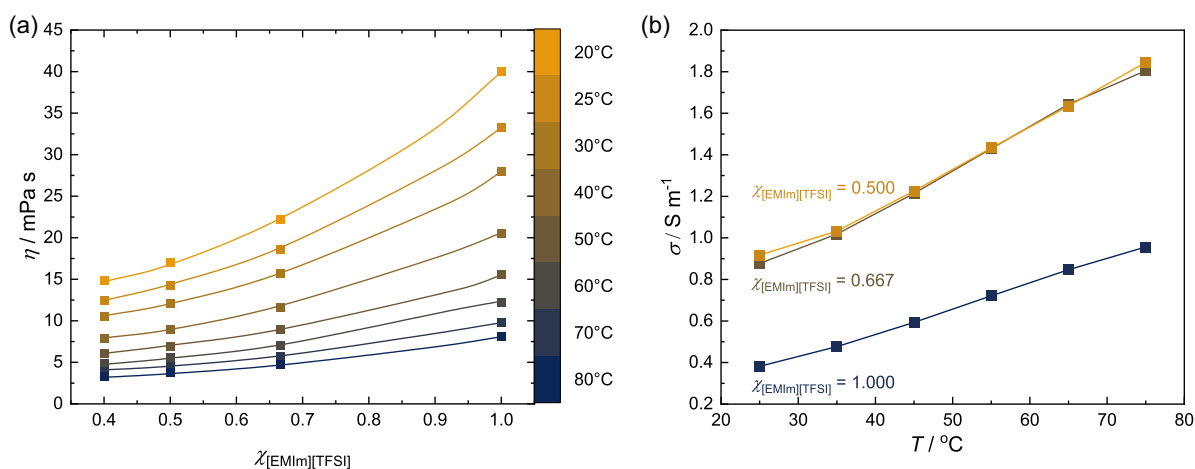


**FIGURE 2** | (a) Dynamic viscosity of the [EMIm][TFSI]:NMA system plotted as a function of molar fraction of [EMIm][TFSI] in NMA and as a function of temperature and (b) conductivity of systems containing [EMIm][TFSI] ( $\chi_{[EMIm][TFSI]}=1.000$ ), [EMIm][TFSI]:NMA 1:0.5 ( $\chi_{[EMIm][TFSI]}=0.667$ ), and [EMIm][TFSI]:NMA 1:1 ( $\chi_{[EMIm][TFSI]}=0.500$ ) as a function of temperature; the lines are drawn as a guide for the eye.

The effect of this structural modification on ionic transport was evaluated by EIS. The temperature-dependent ionic conductivities, determined from the bulk resistance of the systems, are presented in Figure 2b. For all three compositions, conductivity increases continuously with temperature. At 25 °C, neat [EMIm][TFSI] exhibits a conductivity of approximately  $0.38 \text{ S m}^{-1}$ , which increases to  $0.96 \text{ S m}^{-1}$  at 75 °C. In comparison, the eutectic electrolyte increases from approximately  $0.88\text{--}1.80 \text{ S m}^{-1}$ , while the 1:1 mixture increases from approximately  $0.92\text{--}1.84 \text{ S m}^{-1}$  over the same temperature range.

Thus, NMA incorporation nearly doubles the conductivity relative to neat [EMIm][TFSI] at room temperature. The conductivity enhancement is consistent with the pronounced viscosity reduction, indicating that the increased mobility of the liquid facilitates charge transport. However, the conductivity does not increase proportionally with increasing NMA content. Despite its lower viscosity, the 1:1 mixture exhibits only slightly higher conductivity than the eutectic composition throughout the investigated temperature range. This behavior reflects the competing contributions of ion mobility and charge-carrier concentration, in which increasing NMA facilitates molecular motion but simultaneously decreases the fraction of ionic species responsible for charge transport. Consequently, once the eutectic composition is reached, further addition of NMA provides only a marginal conductivity benefit.

To further quantify the temperature dependence of ionic transport, the conductivity data are analyzed using an Arrhenius representation (Figure S5). All three compositions show excellent linear correlations, with  $R^2$  values of 0.995, 0.996, and 0.997 for neat [EMIm][TFSI], the eutectic composition, and the 1:1 mixture, respectively. The corresponding apparent activation energies for ionic transport are 16.110, 12.874, and  $12.403 \text{ kJ mol}^{-1}$ , respectively. Thus, NMA incorporation reduces the apparent activation energy by approximately 20% for the eutectic composition and 23% for the 1:1 mixture relative to neat [EMIm][TFSI].

The lower activation energies indicate that ionic transport requires a lower apparent energetic barrier in the NMA-containing electrolytes. This finding complements the viscosity measurements and supports the interpretation that NMA

modifies the strongly interacting ionic-liquid environment, facilitating molecular and ionic rearrangement. Importantly, the activation energies of the eutectic and equimolar compositions are very similar, differing by only  $0.47 \text{ kJ mol}^{-1}$ , despite their different NMA contents. Together with their nearly identical conductivity values, this indicates that most of the transport benefit associated with NMA incorporation is already achieved at the eutectic composition.

These results establish a clear composition–transport relationship. Addition of NMA substantially reduces viscosity and lowers the apparent activation barrier for ionic transport, resulting in a pronounced increase in conductivity. However, increasing the NMA content beyond the eutectic composition produces only a small additional improvement in conductivity because the gain in ionic mobility is increasingly offset by dilution of the ionic charge carriers. Since higher NMA contents also reduce the electrochemical stability of the electrolyte, as discussed in Section 3.3, the eutectic composition provides the most favorable compromise between low viscosity, efficient ionic transport, and electrolyte stability. This balance is particularly important for PTMA electrodes, where rapid ion transport is required to access the redox-active sites throughout the polymer electrode.

### 3.3 | Electrochemical Stability of the Engineered Electrolytes

While reducing the viscosity of ionic-liquid electrolytes is essential for improving ion transport, this modification should not compromise their electrochemical stability. The electrochemical stability window, therefore, represents a critical parameter in evaluating whether the introduction of NMA preserves the intrinsic advantages of [EMIm][TFSI]. To assess this, linear sweep voltammetry was performed for neat [EMIm][TFSI] and the two selected electrolyte compositions (1:0.5 and 1:1), as shown in Figure 3.

As expected, neat [EMIm][TFSI] exhibits a wide electrochemical stability window of approximately 4.53 V, in excellent agreement with literature values for this ionic liquid [40] (Figure 3a). Introducing NMA progressively narrows the electrochemical

stability window, yielding 4.18 V for the eutectic electrolyte and 3.98 V for the equimolar mixture (Figure 3b,c).

The gradual decrease in electrochemical stability window is consistent with the increasing contribution of NMA to the electrolyte composition. Compared with [EMIm][TFSI], NMA possesses a narrower intrinsic electrochemical stability range and therefore becomes the preferential component undergoing electrochemical oxidation or reduction at the electrode surface. Furthermore, increasing the NMA concentration modifies the local solvation environment. At high ionic-liquid contents, NMA molecules remain strongly coordinated by surrounding ionic species, which partially stabilizes them against electrochemical decomposition [39, 41]. As the NMA concentration increases beyond the eutectic composition, a larger fraction of uncoordinated molecules is expected to be present, making the electrolyte increasingly susceptible to parasitic decomposition reactions.

The composition dependence therefore suggests that the effect of NMA is governed by two competing contributions. At moderate NMA concentrations, modification of the ionic interaction network and the resulting reduction in viscosity enhance ionic mobility. With further NMA addition, however, the decreasing fraction of ionic species limits the benefit obtainable from additional viscosity reduction, while the reduced electrochemical stability becomes increasingly detrimental. The eutectic composition consequently represents a transition between these competing effects, providing most of the transport enhancement associated with NMA incorporation while retaining a sufficiently high ionic-liquid content for electrochemical performance.

Despite this moderate reduction in electrochemical stability, the eutectic electrolyte retains an electrochemical stability window of 4.18 V, which remains considerably wider than the operating potential ranges of most polymer-based electrode materials, including TEMPO radicals [39], quinones [42], and viologens [43, 44].

Consequently, the slight decrease in electrochemical stability window is unlikely to limit practical operation in polymer batteries, whereas the substantial reduction in viscosity is expected to provide significant benefits for reaction kinetics and mass transport.

When considered together with the transport measurements, these results highlight the competing effects introduced by NMA. Increasing the NMA content continuously lowers the viscosity but simultaneously reduces electrochemical stability. The eutectic composition represents the optimum compromise, combining a pronounced improvement in transport properties with an electrochemical stability window sufficiently wide for polymer battery applications.

### 3.4 | Influence of Electrolyte Composition on PTMA Electrochemistry

Having established that the eutectic electrolyte combines low viscosity with a sufficiently wide electrochemical stability window, its influence on the electrochemical behavior of PTMA electrodes is studied by cyclic voltammetry. Since ion transport strongly influences the accessibility of redox-active sites within polymer electrodes, the electrolyte composition is expected to directly affect the electrochemical activation, reaction kinetics, and long-term stability of PTMA.

Figure 4 compares the cyclic voltammograms recorded at  $10 \text{ mV} \cdot \text{s}^{-1}$ . Neat [EMIm][TFSI] initially delivers relatively low specific currents and requires repeated cycling before reaching its maximum electrochemical activity (Figure 4a). This gradual increase indicates slow electrochemical activation of the polymer electrode, which can primarily be attributed to the high viscosity of the ionic liquid. The sluggish ion transport delays electrolyte penetration into the porous PTMA electrode and limits the accessibility of redox-active TEMPO sites during the initial cycles.

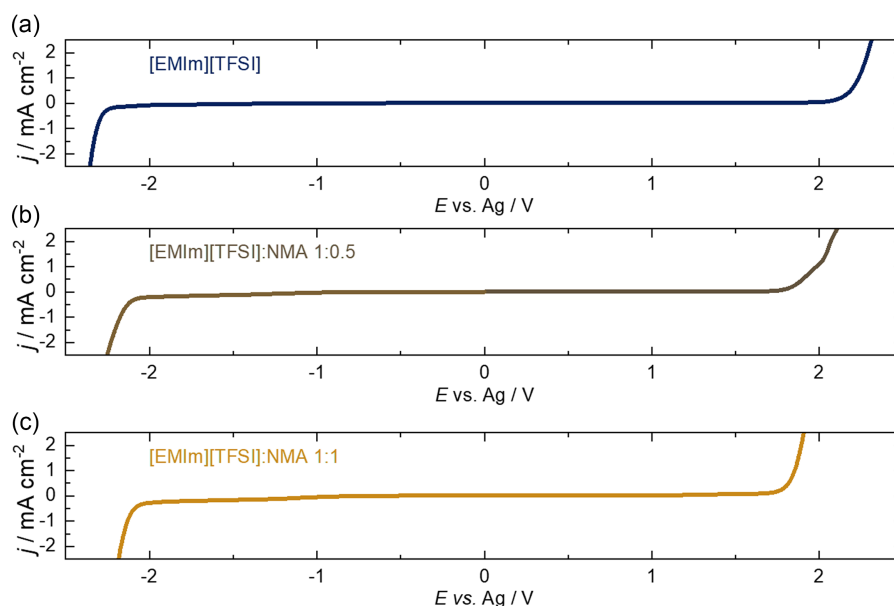


**FIGURE 3** | Linear sweep voltammetry using (a) [EMIm][TFSI] ( $\chi_{[\text{EMIm}][\text{TFSI}]} = 1.000$ ), (b) [EMIm][TFSI]:NMA 1:0.5 ( $\chi_{[\text{EMIm}][\text{TFSI}]} = 0.667$ ), and (c) [EMIm][TFSI]:NMA 1:1 ( $\chi_{[\text{EMIm}][\text{TFSI}]} = 0.500$ ) showing the electrochemical stability window of each mixture with a glassy carbon working electrode.

The addition of NMA significantly accelerates this activation process. The eutectic electrolyte exhibits substantially higher specific currents during the first cycles and reaches a maximum current of approximately  $13.1 \text{ A g}^{-1}$ , considerably exceeding that obtained with neat [EMIm][TFSI] (Figure 4b). The lower viscosity facilitates faster electrode wetting and reduces transport limitations, enabling more efficient utilization of the active material already during the initial cycles. In addition, the redox peaks remain located at more positive potentials, approaching those typically reported for PTMA in conventional electrolytes [18]. This observation suggests that the modified electrolyte environment reduces polarization and improves charge-transfer kinetics at the electrode/electrolyte interface.

Increasing the NMA concentration beyond the eutectic composition initially produces even higher currents; however, these improvements are accompanied by a rapid loss of electrochemical activity upon continued cycling (Figure 4c). This behavior indicates that although additional NMA further enhances ionic transport, it simultaneously promotes degradation processes that progressively outweigh the kinetic benefits. These degradation phenomena may arise from electrochemical decomposition of excess NMA or enhanced dissolution of the polymer in the increasingly molecular solvent environment, ultimately leading to accelerated capacity loss [39].

The influence of transport limitations becomes even more evident at the lower scan rate of  $1 \text{ mV} \cdot \text{s}^{-1}$  (Figure 5). The longer cycling times allow improved electrolyte infiltration into the porous electrode, resulting in significantly faster electrochemical activation for all

electrolytes (Figure 5a–c). Under these conditions, the eutectic electrolyte achieves full activation during the first cycle while maintaining stable current densities throughout the initial cycling (Figure 5b). In contrast, although the equimolar mixture again exhibits relatively high initial currents, rapid degradation is observed, confirming that excessive NMA compromises the long-term electrochemical stability of the PTMA electrode (Figure 5c).

These results demonstrate that the improved electrochemical performance of the eutectic electrolyte originates from a favorable balance between transport enhancement and chemical stability. Lower viscosity accelerates electrolyte wetting and ion transport, enabling faster activation of the polymer electrode, while maintaining sufficient ionic-liquid content suppresses the degradation processes observed at higher NMA concentrations.

### 3.5 | Long-Term Electrochemical Stability

To evaluate the durability of the optimized electrolyte under prolonged operation, freshly assembled cells were subjected to 500 consecutive cyclic voltammetry cycles at  $10 \text{ mV} \cdot \text{s}^{-1}$ . The evolution of the electrochemical response is presented in Figure 6.

Neat [EMIm][TFSI] shows relatively low specific currents throughout the experiment together with a gradual shift in peak potentials, indicating progressive changes at the electrode/electrolyte interface during cycling. The influence of dissolved PTMA on the cell potential drift is discussed in the Figure S6. In contrast, the eutectic electrolyte maintains substantially higher current densities while preserving nearly constant peak positions over the

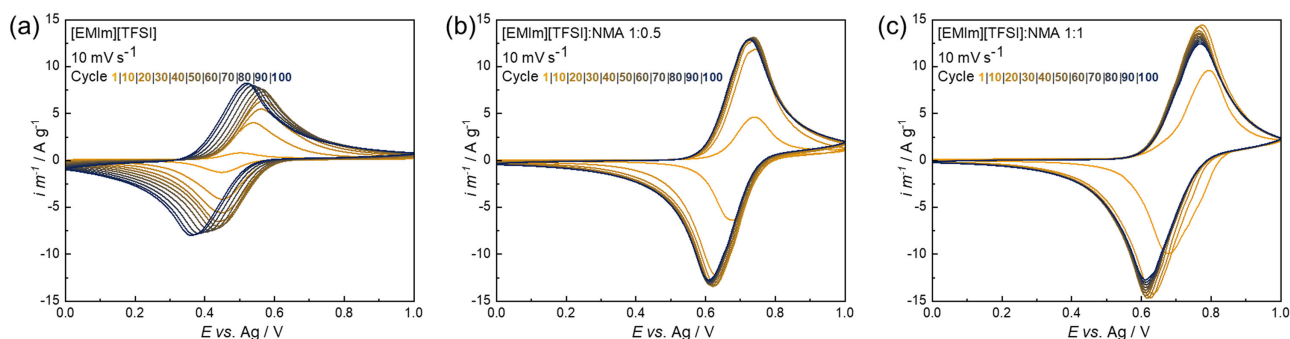


**FIGURE 4** | Half-cell cyclic voltammetry of PTMA working electrode at  $10 \text{ mV} \cdot \text{s}^{-1}$  with (a) [EMIm][TFSI], (b) [EMIm][TFSI]:NMA 1:0.5, and (c) [EMIm][TFSI]:NMA 1:1 as electrolyte for 100 cycles.

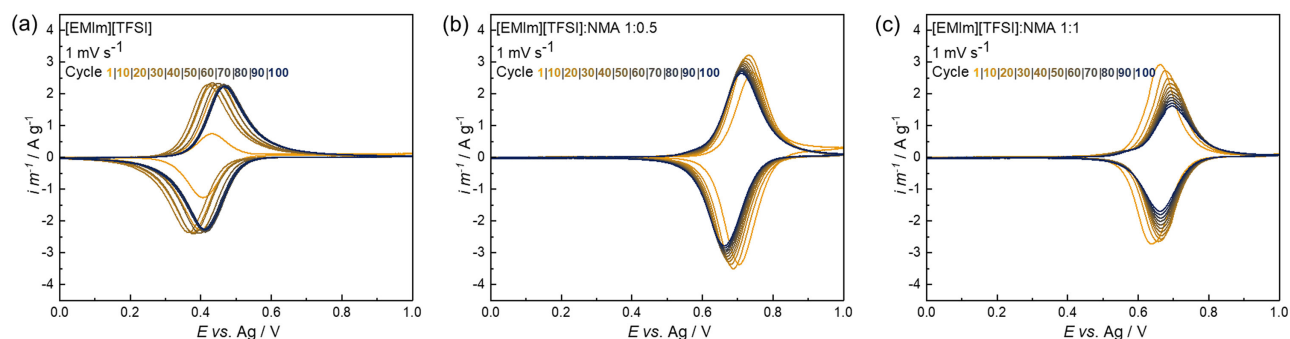


**FIGURE 5** | Half-cell cyclic voltammetry of PTMA working electrode at  $1 \text{ mV} \cdot \text{s}^{-1}$  with (a) [EMIm][TFSI], (b) [EMIm][TFSI]:NMA 1:0.5, and (c) [EMIm][TFSI]:NMA 1:1 as electrolyte for 100 cycles.

entire experiment. After 500 cycles, the loss in specific current amounts to only 31.1%, demonstrating considerably improved electrochemical stability. Furthermore, the equimolar electrolyte exhibits a continuous decrease in peak currents with further cycling, consistent with progressive degradation of the electrochemical system. Although this electrolyte provides the highest initial current densities, its superior transport properties are insufficient to compensate for the reduced electrochemical stability introduced by the higher NMA concentration.

The evolution of the specific charge and discharge capacities (Figure 6d) further illustrates these differences. PTMA cycled in neat [EMIm][TFSI] exhibits the lowest specific capacities throughout the experiment. During approximately the first 80–100 cycles, both the charge and discharge capacities gradually increase until reaching a maximum value of about  $0.05 \text{ mAh} \cdot \text{g}^{-1}$ . This extended activation period indicates that the highly viscous electrolyte requires repeated cycling before sufficient electrolyte penetration and ionic transport pathways are established throughout the porous PTMA electrode. After reaching the maximum capacity, the electrochemical response remains relatively stable for a limited number of cycles before gradually declining toward the end of the 500-cycle experiment. In contrast,

the eutectic electrolyte ([EMIm][TFSI]:NMA = 1:0.5) exhibits capacities of approximately  $0.07 \text{ mAh} \cdot \text{g}^{-1}$  within the first few cycles, indicating that the optimized electrolyte facilitates the immediate utilization of the active material. Following this rapid activation, both charge and discharge capacities remain nearly constant over several hundred cycles, with only a slow and continuous decrease observed during prolonged operation. Even after 500 cycles, the eutectic electrolyte maintains the highest discharge capacity among the three investigated systems, confirming its superior long-term electrochemical stability.

The 1:1 electrolyte exhibits the highest initial capacities, approaching  $0.075\text{--}0.08 \text{ mAh} \cdot \text{g}^{-1}$  during approximately the first 20–30 cycles. However, this initial advantage is rapidly lost. Beginning at roughly 70 cycles, both charge and discharge capacities decrease continuously, and the degradation rate is substantially higher than that observed for the eutectic electrolyte. By the end of the 500-cycle experiment, the remaining capacity is significantly lower despite the excellent initial electrochemical response. The increasing separation between the charge and discharge capacities during prolonged cycling further suggests a gradual loss of electrochemical reversibility. These results clearly demonstrate that the electrolyte providing the highest initial

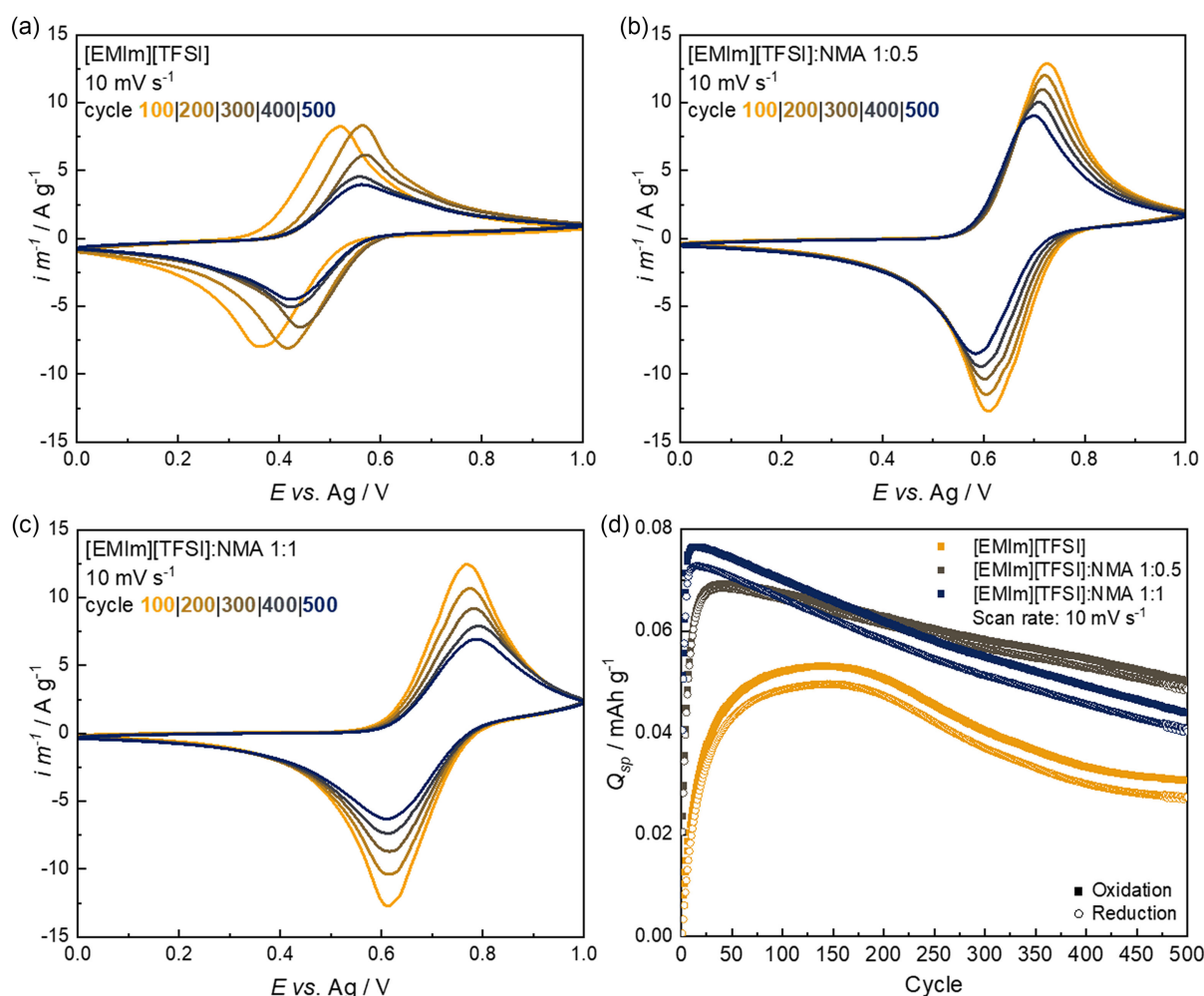


**FIGURE 6** | Half-cell cyclic voltammetry of PTMA working electrode at  $10 \text{ mV} \cdot \text{s}^{-1}$  with (a) [EMIm][TFSI], (b) [EMIm][TFSI]:NMA 1:0.5, (c) [EMIm][TFSI]:NMA 1:1 as electrolyte showing the 100th, 200th, 300th, 400th, and 500th cycle, and (d) specific charge of oxidation and reduction for all three electrolytes for 500 cycles.

capacity is not necessarily the electrolyte delivering the best long-term performance.

To correlate the electrochemical performance with possible structural degradation of the electrodes, postmortem SEM analysis was performed (Figures S7 and S8). The SEM images reveal markedly different degrees of active-material loss depending on the electrolyte composition. Electrodes cycled in neat [EMIm][TFSI] and especially in the 1:1 electrolyte exhibit more pronounced degradation, whereas the eutectic electrolyte largely preserves the electrode morphology. A more detailed discussion of the postmortem SEM analysis is provided in the Supporting Information.

The improved stability of the eutectic electrolyte can be rationalized by considering the balance between transport properties and chemical compatibility. The reduction in viscosity enhances ionic transport and minimizes concentration gradients within the electrode, thereby reducing polarization during cycling. At the same time, maintaining a sufficiently high ionic-liquid fraction suppresses excessive interaction of NMA with the polymer, limiting dissolution of the active material and minimizing electrolyte decomposition. Once the NMA concentration exceeds the eutectic composition, these stabilizing effects are progressively lost, leading to accelerated degradation and reduced electrochemical stability [39, 45].

### 3.6 | Galvanostatic Performance of the Optimized Electrolyte

The influence of electrolyte composition on practical battery performance is finally evaluated under galvanostatic charge-discharge conditions at rates of 10 and 1 C.

At the high rate of 10 C, neat [EMIm][TFSI] delivers only limited charge and discharge capacities because its high viscosity severely restricts ionic transport during the short charging and discharging periods (Figure 7a). In contrast, the eutectic electrolyte achieves 74.5% of the theoretical PTMA capacity at the beginning of cycling and retains 53.5% after 500 cycles, demonstrating a substantial improvement in high-rate capability (Figure 7b). Although the equimolar electrolyte initially exhibits similarly high capacities, it undergoes rapid capacity fading, retaining only a small fraction of its initial capacity after prolonged cycling (Figure 7c).

The superior long-term stability of the eutectic electrolyte becomes even more evident when compared with previously reported ionic-liquid systems. PTMA electrodes employing Pyr<sub>14</sub>TFSI-containing LiTFSI retained approximately 84.7% of their capacity after 200 cycles at 10 C [25], whereas the present eutectic electrolyte achieves a comparable retention of 82.1% over the same cycling interval without the addition of any metal salts. This demonstrates that the performance enhancement originates solely from tailoring the electrolyte structure through eutectic interactions rather than through additional electrochemically active species.

At the lower rate of 1 C (Figure 8), the influence of transport limitations becomes less pronounced because longer charging times allow sufficient electrolyte penetration into the electrode. Consequently, neat [EMIm][TFSI] gradually approaches high discharge capacities after several activation cycles (Figure 8a). Nevertheless, significant capacity fading is observed after approximately 100 cycles, accompanied by Coulombic efficiencies exceeding 100%, indicating the presence of parasitic side reactions. In contrast, the eutectic electrolyte reaches high capacities

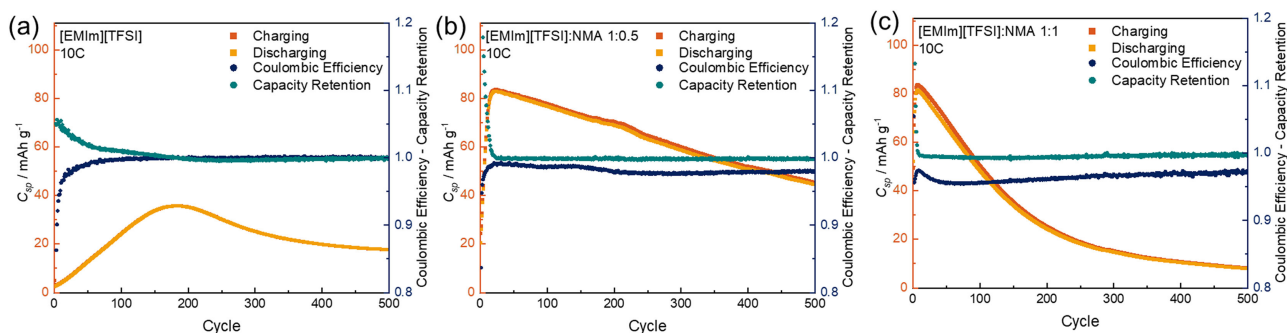


**FIGURE 7** | Half-cell galvanostatic cycling of PTMA working electrode at 10 C rate with (a) [EMIm][TFSI], (b) [EMIm][TFSI]:NMA 1:0.5, and (c) [EMIm][TFSI]:NMA 1:1 as electrolyte for 500 cycles.

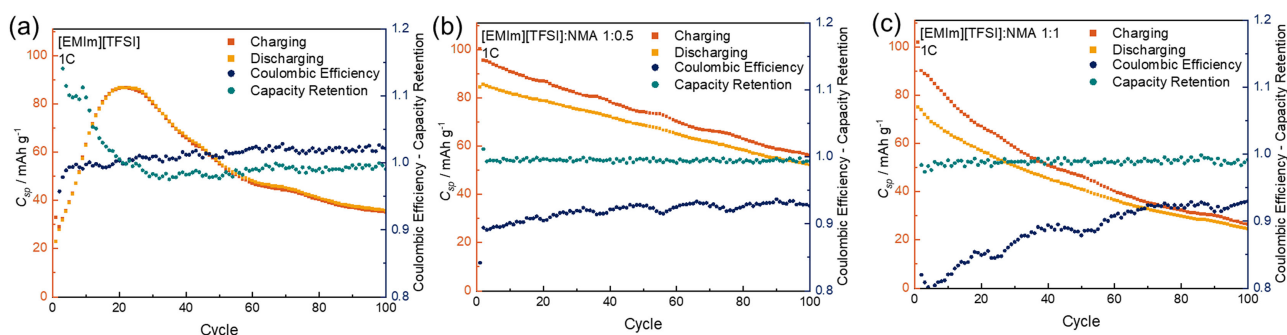


**FIGURE 8** | Half-cell galvanostatic cycling of PTMA working electrode at 1 C rate with (a) [EMIm][TFSI], (b) [EMIm][TFSI]:NMA 1:0.5, and (c) [EMIm][TFSI]:NMA 1:1 as electrolyte for 100 cycles.

immediately after cell assembly and exhibits significantly improved capacity retention while maintaining stable electrochemical behavior (Figure 8b). The equimolar electrolyte again displays inferior cycling stability and lower Coulombic efficiency, confirming that excessive NMA promotes degradation rather than improving battery performance (Figure 8c).

The electrochemical results consistently demonstrate that battery performance is governed by two competing processes introduced by NMA. Hydrogen-bond-mediated restructuring of the electrolyte lowers viscosity and enhances ionic transport, resulting in faster electrode wetting, reduced polarization, and improved utilization of the PTMA-active material. Conversely, excessive NMA decreases the electrochemical stability of the electrolyte and promotes polymer degradation and parasitic side reactions. The eutectic composition represents the optimum balance between these opposing effects, simultaneously providing improved transport properties, sufficient electrochemical stability, and excellent long-term cycling performance. More broadly, these findings demonstrate that DES chemistry can serve as a rational molecular engineering strategy for optimizing ionic-liquid electrolytes, extending the DES concept beyond conventional eutectic solvents toward the design of next-generation electrolytes for sustainable polymer batteries.

## 4 | Conclusion

This study demonstrates that eutectic mixing with NMA can effectively modify the transport properties of [EMIm][TFSI] for PTMA-based organic electrochemistry. The identified [EMIm][TFSI]:NMA = 1:0.5 eutectic composition reduced the viscosity at 20 °C from 40.05 to 22.20 mPa s (45%) and increased the ionic conductivity at 25 °C from 0.38 to 0.88 S · m<sup>-1</sup>. Temperature-dependent conductivity measurements further showed a lower apparent activation energy for ionic transport, decreasing from 16.11 kJ mol<sup>-1</sup> for neat [EMIm][TFSI] to 12.87 kJ mol<sup>-1</sup> for the eutectic electrolyte.

Increasing the NMA content to the 1:1 composition produced only a marginal further increase in conductivity, while substantially reducing electrochemical stability and long-term cycling performance. In contrast, the eutectic composition maintained a sufficiently wide electrochemical stability window and provided stable PTMA electrochemistry over prolonged cycling. These results show that the optimum electrolyte is not determined by transport properties alone but by the balance between ionic mobility, charge-carrier concentration, and electrochemical stability.

Overall, the [EMIm][TFSI]/NMA eutectic composition provides a practical route to reducing the transport limitations of a viscous ionic-liquid electrolyte while retaining its suitability for PTMA-based organic electrochemical systems.

## Acknowledgments

We thank Prof. Dr. Ulrich Schubert (FSU Jena) for providing the PTMA particles for the electrodes used in this study. We also thank Dr. Mohammad Al-Shakran for recording SEM images of the electrodes.

We thank the German Research Foundation (DFG) for funding under Project ID 390874152 (POLiS Cluster of Excellence, EXC-2154), as well as for funding within the priority program SPP 2248 Polymer-based Batteries (Project ID 441209207). Further, support by the Federal Ministry of Education and Research (Bundesministerium für Bildung und Forschung, BMBF) of Germany within the project "OptiABBY" (no. 03XP0531B) is acknowledged.

Open Access funding enabled and organized by Projekt DEAL.

## Funding

This study was supported by Deutsche Forschungsgemeinschaft (390874152 and 441209207) and the Bundesministerium für Bildung und Forschung (03XP0531B).

## Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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### Supporting Information

Additional supporting information can be found online in the Supporting Information section.