Thermophysical and Electrochemical Properties Measurements of Novel Choline-based Ionic Liquids.

THESIS

By

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Under the advisory of

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Abstract

The urgent need for sustainable energy storage propels the search for advanced battery technologies. Traditional lithium-ion batteries with liquid electrolytes pose safety risks like flammability and leakage. Choline-based ionic liquids (ILs) incorporated into gel polymer electrolytes (GPEs) offer a promising solution with the potential to enhance safety, cost-effectiveness, and environmental sustainability. This thesis focuses on the development and optimization of choline-based IL GPEs specifically for lithium battery applications.

Choline is a vital water-soluble nutrient that plays a crucial role in numerous physiological processes including neurotransmitter synthesis, cell membrane integrity, and lipid transport. Its versatility extends to diverse fields like aquaculture and beef production, highlighting its potential for large-scale applications. Although the versatility of choline is very broad, research on its usage in lithium-ion batteries is very limited. Choline-based ionic liquids inherit properties like biocompatibility, biodegradability, and cost effectiveness while offering the tunability needed to tailor electrochemical characteristics. These properties make them promising candidates for electrolytes in advanced battery technologies.

A series of novel choline-ester ILs were synthesized using facile methods, including an acid-base reaction to produce choline hydroxide and subsequent esterification with selected carboxylic acids. Their structures were confirmed by NMR and FTIR. These choline ester ILs exhibited high thermal stability (up to 300°C), non-flammability, high ionic conductivities (up to 60.92 $mScm^{-2}$), high oxidative stability (up to 4.76 V), and impressive lithium-ion transference numbers (up to 0.94). GPEs were fabricated by incorporating these ILs into a PVDF-HFP matrix using a solution casting technique. The fabricated GPEs demonstrated significant safety improvements compared to traditional liquid electrolytes while maintaining electrochemical performance comparable to other ILGPEs systems. The structure-property relationships uncovered, particularly the effect of alkyl chain length on ionic conductivity, offer valuable guidance for further optimization.

This work underscores the potential of choline based ILs as a versatile electrolyte platform. It also highlights the advantages of GPEs for creating safer, high-performance lithium-ion batteries. Further research exploring other choline derivatives, advanced GPE architectures, and detailed battery testing could unlock even greater breakthroughs in energy storage technology. These future research directions could include investigations into bio-derived polymers, the inclusion of fillers for enhanced mechanical and electrochemical properties, and the development of solid-state electrolytes based on choline chemistries.

Keywords

Ionic liquids; gel polymer electrolyte; lithium ion batteries

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Declaration

The investigation presented in this thesis was conducted in the Department of Chemical and Environmental Engineering, University of Nottingham Malaysia (between December 2019 and April 2024). I declare that this work is purely based on my research findings and has not been submitted for any degree to any other institution. I declare that there are no known competing financial interests or personal relationships that could have appeared to influence the work reported in this thesis.

Abbreviations

AC	activated carbon
ACI	acetylcholine iodide
ACN	Acetonitrile
ALIBs	Aqueous lithium ion batteries
BF_4^-	Tetrafluoroborate
BMIM	1-butyl-3-methylimidazolium
BPY	*n*-butylpyridinium
Br^-	bromide
C_2H_4	ethene
C_3H_6	propene
CA	cellulose acetate
CBILs	choline-based ionic liquids
CEI	Cathode Electrolyte Interface
CH_2CH_2OH	hydroxyethyl group
CH_3	methyl group
CH_3COO^-	acetate
ChCl	choline chloride
ChOH	choline hydroxide
Choline 2-methylbutyrate	[Ch][2 mb]
Choline acetate	[Ch][Ac]
Choline glutarate	[Ch][Glu]
Choline isobutyrate	[Ch][Ib]
Choline isovalerate	[Ch][Iv]
Choline malonate	[Ch][Mal]
ChTFSI	choline bis(trifluoromethylsulfonyl)imide
Cl^-	Chloride
CMC	carboxymethyl cellulose
CO_2	carbon dioxide
CV	Cyclic Voltammetry
D_3F	1,3,5-trimethyl-1,3,5-tris(3,3,3-trifluoro propyl)-cyclotrisiloxane
DCS	Differential Scanning Calorimetry
DEC	diethylene carbonate
DESs	Deep Eutectic Solvents
DFT	Density Functional Theory
DMC	dimethyl carbonate
DME	1,2-dimethoxyethane
DMSO	dimethyl sulfoxide

DOL	1,3-dioxolane
DSSCs	Dye-sensitized solar cells
E_{AL}	anodic limits
EC	ethylene carbonate
ECL	cathodic limits
EDLC	electric double-layer capacitor
EG	ethylene glycol
EIS	Electrochemical Impedance Spectroscopy
EMC	ethyl methyl carbonate
EMIM	1-ethyl-3-methylimidazolium
EmimI	1-etyl-3-methylimidazolium iodide
EMIM-TFSI	1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide
ESW	electrochemical stability window
ETGs	Eutecto Gels
EW	wide electrochemical window
FEC	fluorinated ethylene carbonate
Fo	formate
FTIR	Fourier Transform Infared Relecteance
G	glycerol
GCD	Galvanostatic Charge Discharge
GPEs	Gel Polymer Electrolytes
H_2	hydrogen
HBA	hydrogen bond acceptor
HBD	hydrogen bond donor
HEC	2-hydroxyethyl cellulose
HF	hydrogen fluoride
I^-	iodide
ILs	Ionic Liquids
IPCC	The Intergovernmental Panel on Climate Change
IPMCs	ionic polymer-metal composites
ISC	short-circuit current
KI	potassium iodide
LA	lactic acid
LBO/Li_3BO_3	tri-lithium borate
LCO	lithium cobalt oxide
LEC	liquid electrolyte chemistry
LEDC	lithium ethylene dicarbonate
LFP	lithium iron phosphate
Li_2SO_4	lithium sulfate
LIBs	lithium ion batteries
LiCl	lithium chloride
LiFSI	lithium bis(fluorosulfonyl)imide
$LiNO_3$	lithium nitrate

$LiPF_6$	Lithium hexafluorophosphate
LiTFSI	Lithium bis(trifluoromethanesulfonyl)imide
LLTO	Li0.33La0.557TiO3
LLZO	$Li_7La_3Zr2O_{12}$
LLZTO	$Li_{6.4}La_3Zr_{1.4}Ta_{0.6}O_{12}$
LMBs	lithium metal batteries
LSV	Linear Sweep Voltammetry
LTO	lithium titanate
MAc	(N-methylacetamide)
MC	methylcellulose
MD	molecular dynamics
MFA	methyl difluoroacetate
MPEGA	polyethylene glycol methyl ether acrylate
MPII	1-methyl-3-propyl imidazolium
MPY	n-methylpyridinium
N^+	Nitrogen ion
NMR	Nuclear Molecular Resonance
NOAA	National Oceanic and Atmospheric Administration
O_2	oxygen
OCP-POSS	polyhedral oligomeric silsesquioxane
OCP-POSS	octa-(3-chloropropyl)-polyhedral oligomeric silsesquioxane
OMIM	1-octyl-3-methylimidazolium
PA	propionic acid
PAA	poly(acrylic acid)
PC	propylene carbonate
PCE	power conversion efficiency
PDMS	Poly(dimethylsiloxane)
PDT	photodynamic therapy
PEGDA	poly (ethyleneglycol) diacrylate
PEGMA	poly(ethylene glycol methacrylate)
PEO	polyethylene oxide
P-ETGs	polymetric eutectogels
PF6-	hexafluorophosphate
PILBCP-TFSI	polymerized ionic liquid diblock copolymer
PILs	poly(ionic liquids)
PLLA	poly-l-lactic acid
PMMA	Polymethyl methacrylate
PPC	poly(propylene carbonate)
PS	polystyrene
PVA	polyvinyl alcohol
PVDF	Polyvinylidene fluoride
PVDF-HFP	poly(vinylidene fluoride-co-hexafluoropropylene)

$PYR_{13}TFSI$	*n*-methyl-*n*-propylpyrrolidinium bis(trifluoromethanesulfonyl)imide
RTIL	room temperature ionic liquid
SA	salicylate
SCEs	solid composite electrolytes
SEI	solid electrolyte interphase
SEM	scanning electron microscopy
SPEs	Solid polymer electrolytes
SPI	soy protein isolate
SSEs	solid-state electrolytes
TBA	tetrabutylammonium
TBP	tetrabutylphosphonium
Tc	crystallization points
TEGDA-BA	triethylene glycol diacetate-2-propenoic acid butyl ester
TFSI	bis(trifluoromethylsulfonyl)imide
Tg	glass transition temperature
TGA	Thermogravimetric Analysis
THF	tetrahydrofuran
Tm	melting points
T_{onset}	onset decomposition temperatures
TrFE	Trifluoro ethylene
Triflate	trifluoromethanesulfonate
U	urea
VOC	open-circuit voltage
VTF	Vogel-Tamman-Fulcher
WIS	water-in-salt
WiSIL	water-in-salt ionic liquid
XPS	X-ray photoelectron spectroscopy
β-Ala	beta-alanine

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Chapter 1: Introduction

1.1 Background of the Study

According to the National Oceanic and Atmospheric Administration (NOAA), 2023 is the warmest year on record (LINDSEY and DAHLMAN, 2024) (better illustrated in Figure 1). The occurrence of global warming, driven by climate change, threatens biodiversity, ecosystems, and ecosystem services. The Intergovernmental Panel on Climate Change (IPCC) predicts a substantial increase in global temperatures by 2100 (Anon and Kumar, 2021)

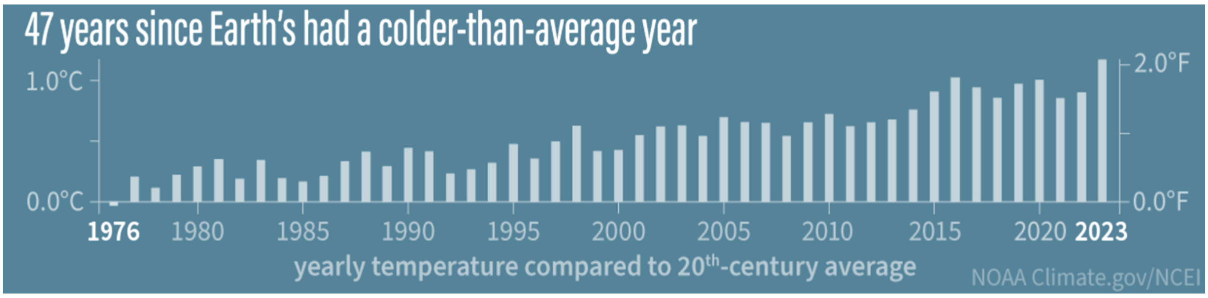


Figure 1. The bar graph visually represents the annual global temperatures spanning from 1976 (on the left) to 2023 (on the right) in relation to the average temperature for the years 1901-2000. In 1976 (on the far left), it was noted as the final year when temperatures were lower than the average for the 20th century. Conversely, 2023 (far right) marked a new milestone by being identified as the warmest year on record (LINDSEY and DAHLMAN, 2024).

These are the consequences caused by human activities, such as burning fossil fuels, deforestation, industrial emissions, and agricultural practices. These activities intensify greenhouse gas emissions, resulting in imbalances in natural resource exploitation, changes in global temperature, and precipitation patterns (Ebi, 2020; McGuire, 2021). The current mitigation or fight against global warming is a multi-faceted approach focused on reducing greenhouse gas emissions and enhancing natural carbon sinks. A core strategy involves transitioning from fossil fuels like coal and oil towards renewable energy sources. This includes harnessing the power of the sun through solar energy, utilizing wind turbines to generate electricity, tapping into the Earth's heat with geothermal energy, and carefully employing sustainable biomass for power generation (Cano *et al.*, 2022). The most significant renewable energy source globally is hydroelectric energy, which has historically been a major contributor to renewable energy production (Berga, 2016). However, the drawback of hydroelectricity as a renewable energy source is its potential negative impact on the environment and local populations. The construction of hydroelectric power plants can lead to changes in land cover, fragmentation, degradation, and loss of tropical forests, altering local ecosystems and affecting biodiversity (Velastegui-Montoya, de Lima and Herrera-Matamoros, 2022). Additionally, the installation of hydroelectric power complexes in regions like the Amazon has been shown to cause land cover changes and alter local population dynamics, highlighting the environmental and social challenges associated with large-scale hydroelectric projects (Velastegui-Montoya, de Lima and Herrera-Matamoros, 2022). These impacts underline the importance of considering the environmental consequences of hydroelectricity generation alongside its benefits in the transition to renewable energy sources. In recent years, other sources, such as wind and solar energy, have been rapidly increasing their share in the electrical grids, especially in Asia. Rapid strides are evident in countries such as China and India, which have demonstrated an annual growth rate surpassing 30% in the solar and wind sectors (Sokulski *et al.*, 2022; Hassan *et al.*, 2024). Solar energy is emphasized as a crucial renewable energy source in certain regions due to its compatibility with local climatic conditions and its role in sustainable economic development (Sofiu, Sofiu and Gashi, 2022). However, solar and wind have their drawbacks, which are that their intermittency leads to power generation fluctuations due to weather variations, day-night cycles, and regional differences (Lustfeld, 2021; Kexuan Wang, Li, *et al.*, 2023). This intermittency necessitates using energy storage systems to ensure continuous power supply, adding extra costs to the overall renewable energy infrastructure

(Awad *et al.*, 2022). Additionally, the lack of linearity in solar energy reception by panels, caused by Earth-Sun relative displacement, can be a disadvantage to consider in optimizing energy capture (Gómez-Uceda *et al.*, 2020). Overcoming these challenges through advancements in energy storage technologies and system optimization is crucial for maximizing the efficiency and reliability of solar and wind renewable energy sources.

The inherent intermittency of solar and wind power highlights the urgent need for reliable and efficient energy storage solutions. Lithium ion batteries have emerged as a leading technology for addressing this challenge. Their capability to store energy for later use allows for a more consistent energy supply from renewable sources. However, like any solution, lithium ion batteries come with their own set of drawbacks that need to be addressed for widespread adoption.

Lithium ion batteries are widely recognized for their high energy and power density, lack of memory effect, and extensive use across various applications (Wang *et al.*, 2014; TANG, YIN and QIU, 2017; M. Li *et al.*, 2021). These batteries have become a preferred choice for energy storage in electric vehicles, portable electronics, and power grid systems (Jagatap, 2019; L. Zhang *et al.*, 2020). Continuous advancements in lithium ion battery technology have resulted in enhancements in energy density, cycle life, and cost reduction, solidifying their position as a top option for many energy storage needs (Yang, Lu and Qi, 2013; N. Li *et al.*, 2015). Research efforts have been concentrated on improving lithium ion batteries application through various methods, such as integrating photocatalysts for direct solar energy storage (Zhou, 2016), developing new electrode materials like MX (M=Ge, Sn; X=S, Se) sheets (Zhang *et al.*, 2021), and exploring innovative strategies like the "Anchor-Turbo" approach for high-performance lithium-sulfur batteries (Stroe *et al.*, 2014). Studies have also delved into the state-of-health diagnosis of lithium ion batteries using electrochemical impedance spectroscopy (Lu and Wang, 2022) and the design of remote monitoring systems to monitor the operational status of lithium batteries (G. Liu *et al.*, 2022). Despite the remarkable progress and extensive applications of lithium ion batteries, certain inherent limitations pose challenges that have spurred significant research efforts. One significant drawback of lithium batteries is safety concerns, particularly related to the flammability of the liquid electrolytes commonly used in lithium ion batteries (Srour, Rouault and Santini, 2013; Park *et al.*, 2020). These electrolytes are highly flammable and reactive with electrode materials, posing safety risks such as fire hazards and potential leakage issues (Park *et al.*, 2020). Additionally, the high reactivity of these electrolytes can lead to safety issues caused by overcharging, over-discharging, short circuits, and production defects (Srour, Rouault and Santini, 2013; Ghaeminezhad and Monfared, 2022). The safety risks associated with traditional lithium ion batteries have prompted a search for alternative electrolyte materials. One promising class of materials is ionic liquids.

1.1 Ionic Liquids (ILs)

Ionic liquids are a class of compounds that have gained significant attention due to their unique properties. These liquids consist entirely or almost entirely of ions, distinguishing them from conventional molecular liquids (Sánchez-Ramírez *et al.*, 2017). The traditional definition of ionic liquids is based on their physical state, with a key criterion being that they are salts that are liquid at or below 100 °C (Ganjoo *et al.*, 2022). However, the definition of ionic liquids has been a topic of debate, with some arguing that the term is somewhat arbitrary and may not fully capture the diverse range of properties exhibited by these compounds (Hayes, Warr and Atkin, 2015). While the conventional definition emphasizes the melting point criterion, it is important to note that ionic liquids can exhibit a wide range of properties beyond just being liquid at room temperature. For instance, some ionic liquids may exist as solids at room temperature, challenging the notion of them being purely liquid compounds (Stocker, Healy and Ferguson, 2020). Additionally, the definition of ionic liquids has evolved to encompass variations such as poly(ionic liquids) (PILs), which are macromolecules containing ionic groups characteristic of classical ionic liquids (W. Xu *et al.*, 2015).

Ionic liquids are a diverse class of salts that remain liquid at room temperature. Their unique properties stem from their composition: bulky organic cations and a variety of organic or inorganic anions (Gupta and K. Singh, 2021). With its poorly coordinated ions, this structure grants ILs characteristics such as negligible vapor pressure, a wide liquid range, non-flammability, high conductivity, and versatile solvent properties (Ramajo *et al.*, 2021). A key advantage of ILs is their tunability – by carefully selecting cations and anions, researchers can design ionic liquids with tailored properties for specific applications (Sasi, Sarojam and Devaki, 2016). The common cations found in ionic liquids include (Figure 2):

- Imidazolium: Examples include 1-ethyl-3-methylimidazolium (EMIM), 1-butyl-3-methylimidazolium (BMIM), and 1-octyl-3-methylimidazolium (OMIM) (Nazet *et al.*, 2015).
- Pyridinium: Such as *n*-butylpyridinium (BPY) and *n*-methylpyridinium (MPY) (Chaban and Fileti, 2016).
- Ammonium: Quaternary ammonium cations like tetrabutylammonium (TBA) (Biczak *et al.*, 2017).
- Phosphonium: Such as tetrabutylphosphonium (TBP) (Plaquevent *et al.*, 2008).
- Sulfonium: For example, trialkylsulfonium (Jungen and Chen, 2018).

ILs can be further customized by pairing these cations with various anions, including:

- Halides: Chloride (Cl^-), bromide (Br^-), iodide (I^-) (Llewellyn Lancaster *et al.*, 2002).

- Fluorinated Anions: Tetrafluoroborate (BF_4^-), hexafluorophosphate (PF_6^-), bis(trifluoromethylsulfonyl)imide (TFSI), trifluoromethanesulfonate (Triflate) (Mukaromah *et al.*, 2023).
- Other Common Anions: Dicyanamide, thiocyanate, nitrate, acetate, alkyl sulfates (Mondal and Balasubramanian, 2015).

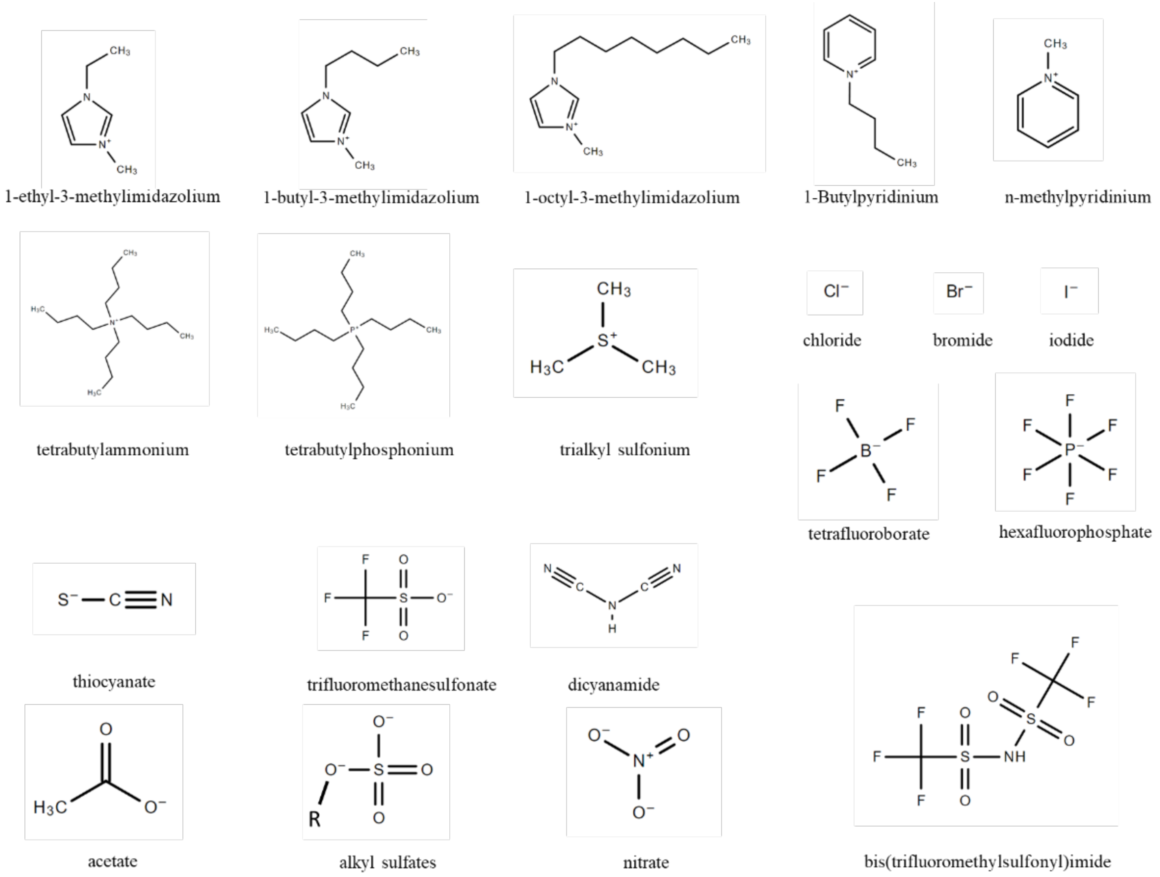

1-ethyl-3-methylimidazolium
1-butyl-3-methylimidazolium
1-octyl-3-methylimidazolium
1-Butylpyridinium
n-methylpyridinium
tetrabutylammonium
tetrabutylphosphonium
trialkyl sulfonium
chloride
bromide
iodide
tetrafluoroborate
hexafluorophosphate
thiocyanate
trifluoromethanesulfonate
dicyanamide
bis(trifluoromethylsulfonyl)imide
acetate
alkyl sulfates
nitrate

Figure 2. Chemical structure of common cations and anions of ILs.

This remarkable tunability of ionic liquids, achieved through both cation-anion combinations and ion modifications, has enabled the creation of task-specific ILs addressing a wide range of challenges in fields like catalysis (Amarasekara, 2016; Wu *et al.*, 2018; Bartlewicz *et al.*, 2020), materials science (García, Atilhan and Aparicio, 2015; THACH *et al.*, 2019; Galvan *et al.*, 2021), green chemistry (Banan *et al.*, 2017; Costa, Forster-Carneiro and Hallett, 2023), separation and purification (Ao *et al.*, 2017; Turgis *et al.*, 2018; Pathak *et al.*, 2023), and biotechnology and pharmaceuticals (Mouradzadegun *et al.*, 2017; Abadast, Mouradzadegun and Ganjali, 2020; Manjarrez, Clark and Fieser, 2023). Notably, the drive for safer and more sustainable solutions, evident across various fields, extends to the realm of energy storage. Their high thermal stability, low flammability, and non-volatility significantly enhance battery safety (Q. Wang *et al.*, 2020). ILs also offer high ionic conductivity for improved charge transfer (Q. Wang *et al.*, 2020) and are highly customizable for optimized battery performance (Verma, Ebenso and Quraishi, 2018). Additionally, their environmentally friendly nature makes them a sustainable choice for energy storage systems (Saha, 2021). While ionic liquids offer significant advantages, researchers are actively addressing potential limitations such as the synthesis and purification of specialized ionic liquids can be expensive (Hernández-Beltrán *et al.*, 2019; THACH *et al.*, 2019; Gholami, Pourfayaz and Maleki, 2020), especially for large-scale applications, hindering their widespread commercial adoption. Additionally, many ionic liquids exhibit high viscosity (Philippi *et al.*, 2019), viscosity of ionic liquids can impede their practical use in certain applications, especially those requiring fast diffusion or flow of the liquid (Vo *et al.*, 2019). The high viscosity of ionic liquids can also affect their performance in processes such as energy storage and CO_2 capture (Orhan, Ume and Alper, 2017). Maintaining high purity is another challenge, as impurities can affect the stability and performance of ionic liquids (Schmeisser and Van Eldik, 2009), further adding to production costs. Moreover, while many ILs are less toxic than traditional electrolytes, the long-term environmental effects of some ionic liquids are not fully understood, necessitating ongoing research into biodegradable ILs (Deferm *et al.*, 2017). Lastly, while research has made significant strides, the sheer number of potential ionic liquid combinations (estimated at 10^18) (Aparicio, Atilhan and Karadas, 2010) poses a challenge to fully understanding their complex interactions with various materials. To date, fundamental properties have been measured for only a small fraction of these possible ionic liquids. Even the most extensive databases, like the IL Thermo database, contain data only just 2332 pure ionic liquid systems (National Institute of Standards and Technology, 2014), often limited to atmospheric pressure and ambient temperature. These limitations highlight the importance of continued research into suitable ionic liquids for energy storage applications. By carefully designing and optimizing ILs, it is possible to unlock their full potential while addressing the challenges that hinder their broad adoption.

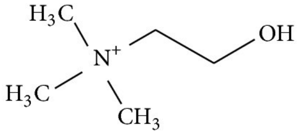
1.2 Choline Based Ionic Liquids

Despite the compelling advantages of ionic liquids, certain drawbacks exist as described in the previous section. Fortunately, the inherent tunability of ILs provides a pathway for overcoming these limitations. By strategically modifying or choosing their structure, ILs can be engineered as potential lithium battery electrolytes, choline based ionic liquids, for instance, can be potentially strong candidates. Choline is a vital, water-soluble nutrient that plays a crucial role in various physiological processes throughout the human lifespan. Its functions range from supporting the synthesis of the neurotransmitter acetylcholine, essential for memory and cognition, to maintaining cell membrane integrity, lipid transport, and methyl-group metabolism (Wiedeman *et al.*, 2018; Goh, Cheam and Wang, 2021).

Moreover, choline plays a significant role in both the understanding of diseases and in medical diagnosis. Aberrant choline metabolism, characterized by elevated levels of choline-containing compounds, is recognized as a metabolic hallmark of cancer development and progression (Gokhale and Xie, 2021). Studies continue to explore choline's role in various health conditions, including liver disease, cardiovascular health, and fetal alcohol spectrum disorders (Shahsavari, D’Occhio and Al Jassim, 2016; Zeisel, Klatt and Caudill, 2018; Ernst *et al.*, 2022). In the diagnostic realm, choline PET imaging offers valuable insights, aiding in procedures like the preoperative localization of hyperfunctioning parathyroid glands(Ferrari *et al.*, 2021).

Beyond its essential role as a nutrient or biomarker in the human body, choline finds applications in various other areas. One notable application is aquaculture, where choline plays a crucial role in lipid metabolism for fish. Studies have shown that choline deficiency in aquafeed can lead to compromised fish health and growth performance (Liu *et al.*, 2021). Additionally, choline is utilized in the beef production system, particularly in the Southern Great Plains, where beef consumption contributes to the human diet. The beef production system in this region produces more human-absorbable choline in the human diet than is consumed in the beef supply chain (Lancaster *et al.*, 2022).

The wide-ranging roles of choline highlight its remarkable versatility. From supporting essential human physiological functions to emerging applications in diagnostics, aquaculture, and beef production, choline demonstrates its importance across diverse fields. Its widespread adoption and versatile functionality make large-scale production strategically and economically viable. Choline can be industrially produced through a specific process involving the reaction of trimethylamine, hydrochloric acid, and ethylene oxide (Brzęczek-Szafran, Siewniak and Chrobok, 2023). This industrial method allows for the mass production of choline chloride, a common form of choline used in various industries, including the poultry sector, as a growth accelerator in chicken feed (Alonso *et al.*, 2016). This production method of choline chloride from these raw materials is cost-effective, simple, and suitable for large-scale manufacturing (K. Oklu, C. Matsinha and C.E. Makhubela, 2020). Furthermore, choline can be derived from various sources, including animal waste and legumes. While choline chloride can theoretically be extracted directly from biomass such as animal waste or legumes, the current industrial production primarily relies on the above reaction process (Brzęczek-Szafran, Siewniak and Chrobok, 2023). This method ensures a consistent and reliable supply of choline for various applications. The ability to produce on a large scale fuels significant research interest in utilizing choline-based materials, highlighting the potential for innovative applications built upon this essential molecule.



Choline

Choline-based ionic liquids are a class of quaternary ammonium ionic liquids (Miao, Atkin and Warr, 2022) that have garnered significant attention due to their various applications and unique properties. Ionic liquids are classified as choline-based if they contain a positively charged ion (cation) derived from the molecule choline. Choline ($C_5H_{14}NO$) has a recognizable core structure consisting of a nitrogen atom (N^+) bonded to three methyl groups (CH_3) and a hydroxyethyl group (CH_2CH_2OH) (X. Li *et al.*, 2022). A choline-based ionic liquid's anion (negatively charged part) can vary widely. It might be simple anions like chloride (Cl^-), acetate (CH_3COO^-), or more complex organic anions (Liu *et al.*, 2018). The anion paired with the choline cation influences the overall properties of the ionic liquid. Studies have also shown that choline-based ionic liquids exhibit enhanced biocompatibility due to their composition derived from natural resources, such as amino acids, which are known for their biodegradability (Tarannum, Rao and Fathima, 2018; Moosavi, Banazadeh and Torkzadeh, 2019; Sivapragasam *et al.*, 2019). These properties make choline-based ionic liquids promising candidates for environmentally friendly and sustainable applications where biocompatibility and environmental impact are crucial.

The applications for choline-based ionic liquids are vast and continue to expand. For instance, they are explored for drug delivery, protein stabilization, and antimicrobial agents in the biomedical field. Choline-based ionic liquids have been extensively studied for their role as solubility enhancers in drug delivery systems, potentially improving drug delivery efficiency

(Caparica *et al.*, 2018). They also offer exciting possibilities in green chemistry, acting as solvents for cleaner reactions and processes (Yiin *et al.*, 2021). Choline-based ILs can even play a role in biomass processing, helping break down plant materials to produce biofuels (Ocreto *et al.*, 2022). Within materials science, they enable the creation of novel materials with unique properties (Wysokowski *et al.*, 2023). Other potential applications include areas like CO_2 capture (Zhang, Ji and Lu, 2018).

Choline was chosen for this study on ionic liquids for gel polymeric electrolytes in lithium-ion batteries due to its numerous advantages in sustainability and performance. Choline, a quaternary ammonium compound, is readily available from renewable sources like plant oils and animal tissue, making it an eco-friendly alternative compared to other traditional ionic liquids (Alhajj *et al.*, 2020). Additionally, its biodegradability and lower toxicity make it a more appealing choice, aligning with the principles of green chemistry. Choline's molecular structure allows it to form stable ionic liquids, and its ability to fine-tune properties like viscosity, conductivity, and thermal stability (Miao, Atkin and Warr, 2022) makes it highly suitable for lithium-ion battery applications .

Economically, choline-based ionic liquids offer a cost-effective solution due to the widespread availability of choline chloride, a common and inexpensive feed supplement used in agriculture (Alonso *et al.*, 2016). The scalability of choline production ensures that it can be manufactured at an industrial level at a lower cost than more complex ionic liquids, which often require expensive precursors or involve labor-intensive synthesis processes. This affordability adds to its appeal as a feasible alternative for use in large-scale energy storage systems like lithium-ion batteries.

From an environmental standpoint, choline-based ionic liquids provide significant advantages. Their low toxicity and biodegradability mean they are less harmful to ecosystems compared to other conventional ionic liquids, many of which are non-biodegradable and toxic to aquatic life (Radošević *et al.*, 2015). By minimizing environmental impact during production, handling, and disposal, choline-based alternatives contribute to a more sustainable approach to battery technology. Their reduced environmental footprint aligns with the growing demand for greener energy storage solutions.

Safety is another crucial factor in the selection of choline. Choline-based ionic liquids generally exhibit low flammability and good thermal stability (Gadilohar and Shankarling, 2017; Tarannum, Rao and Fathima, 2018), which are critical for safe operation in lithium-ion batteries, especially under varying temperature conditions. Their favorable safety profile is further supported by low toxicity compared to other ionic liquids containing fluorine-based anions, which can be corrosive and harmful (Calandra *et al.*, 2021). This makes choline a safer option for both manufacturing and disposal, addressing key safety concerns associated with the use of lithium-ion batteries.

In comparison to other materials, such as imidazolium-based ionic liquids or those with fluorinated anions, choline offers a safer, more sustainable, and cost-effective alternative. While imidazolium ionic liquids provide high conductivity and stability, they are often more expensive and environmentally harmful (de Jesus and Maciel Filho, 2022). Similarly, fluorinated anions, while offering excellent electrochemical properties, pose greater environmental risks due to their persistence and potential to release toxic by-products (Nam *et al.*, 2024). Choline-based ionic liquids, therefore, offer a balanced approach, combining sustainability, safety, and economic feasibility in the development of lithium-ion battery electrolytes.

However, it's important to acknowledge that choline based ionic liquids, while promising, don't solve every challenge within lithium battery technology. One persistent issue about only using ionic liquid as electrolyte is the potential for electrolyte leakage, which poses safety and performance concerns. To address this, researchers are exploring solid-state electrolytes (SSEs) and quasi-solid-state electrolytes (also known as gel polymer electrolytes) (Xu *et al.*, 2020). These electrolyte systems offer the potential to significantly reduce leakage risks, enhancing the safety and reliability of lithium batteries.

1.3 Gel Polymer Electrolytes (GPEs)

Gel Polymer Electrolytes (GPEs) are an innovative class of materials that function as electrolytes in electrochemical devices like batteries and supercapacitors. They are formed by immobilizing a liquid electrolyte solution within a three-dimensional polymer matrix. This hybrid structure offers several advantages over traditional liquid or solid electrolytes. Firstly, GPEs significantly enhance safety by reducing the risk of electrolyte leakage and flammability (Qiao *et al.*, 2020). Secondly, they maintain high ionic conductivity, comparable to liquid electrolytes, ensuring the rapid transport of ions necessary for efficient device performance(Baskoro, Wong and Yen, 2019). Thirdly, the polymer matrix provides mechanical stability and flexibility, allowing GPEs to conform to various device shapes and withstand bending or deformation(Babu *et al.*, 2021). The common polymers used in GPEs include polyethylene oxide (PEO), poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP), and poly(acrylic acid) (PAA). PEO is favoured for its high ionic conductivity and compatibility with battery production processes (Yanan Wang *et al.*, 2019). PVDF-HFP is valued for its versatility, security, and reliability in various battery applications (K. Luo *et al.*, 2021). PAA is known for its low-temperature tolerance and safety contributions to rechargeable zinc-air batteries (Cui *et al.*, 2023). These polymers are crucial for enhancing ionic conductivity, stability, and safety in GPEs for batteries, such as lithium ion and zinc-air batteries (Chen *et al.*, 2022). In summary, GPEs effectively bridge the gap between liquid and solid-state electrolytes, combining their strengths while mitigating their limitations.

1.4 Problem Statement

The increasing effects of global warming and climate change highlight the urgency of transitioning away from fossil fuels towards renewable energy sources. While solar and wind energy hold great potential, their inherent intermittency due to factors like weather and geographic limitations necessitates the development of reliable energy storage solutions. Lithium ion batteries have emerged as a leading technology for energy storage; however, traditional batteries with liquid electrolytes pose safety risks related to flammability and potential leakage. Ionic liquids offer a potential alternative, but some remain costly, lack biodegradability, and often have limited data on their key thermophysical and electrochemical properties. Choline-based ionic liquid gel polymer electrolytes (GPEs) present a promising approach to address these challenges, offering the potential for enhanced safety, cost-effectiveness, and sustainability, but require systematic study to optimize their performance in battery applications.

1.5 Objectives

Traditional lithium ion batteries with liquid electrolytes suffer from safety concerns such as flammability and leaking. Ionic liquids offer a potential solution but can be expensive and lack biodegradability. Choline-based ionic liquids, particularly those derived from choline, are a promising subclass due to their potential for cost-effectiveness, biodegradability, and tailored properties. However, the specific thermophysical and electrochemical properties of choline based ILs remain largely unexplored. To unlock their potential in battery applications, the following objectives were pursued:

1. To develop facile synthesis methods for choline based ionic liquid.
2. To synthesis choline based ionic liquids with bio-inspired organic acids and identify potential candidate of choline based ionic liquids for the application in lithium battery.

3. To characterise the thermophysical (viscosity and thermal stability) and electrochemical (ionic conductivity, electrochemical stability window, transference number) properties of the synthesized novel choline based ionic liquids using established techniques such as viscometry, Thermogravimetric Analysis (TGA), Differential Scanning Calorimetry (DCS), Cyclic Voltammetry (CV), Electrochemical Impedance Spectroscopy (EIS), Linear Sweep Voltammetry (LSV), Transference number, Galvanostatic Charge Discharge (GCD) and flammability test.
4. To fabricate GPEs using the choline based ILs within a PVDF-HFP matrix and investigate the effect of IL structure on GPEs performance.
5. Compare the properties and battery performance of the choline-based IL GPEs to benchmark GPE systems, hypothesizing that the choline-ester ILs will enhance safety and maintain comparable electrochemical performance.

1.6 Novelty of This Work

While choline-based ionic liquids (ILs) are indeed not a new research area, the novelty of this work lies in the specific application and development of choline-based ionic liquids for gel polymeric electrolytes in lithium-ion batteries. This study introduces a unique combination of choline-based ionic liquids with gel polymer matrices, creating a new class of gel polymer electrolytes (GPEs) that exhibit enhanced performance in terms of ionic conductivity, electrochemical stability, and safety.

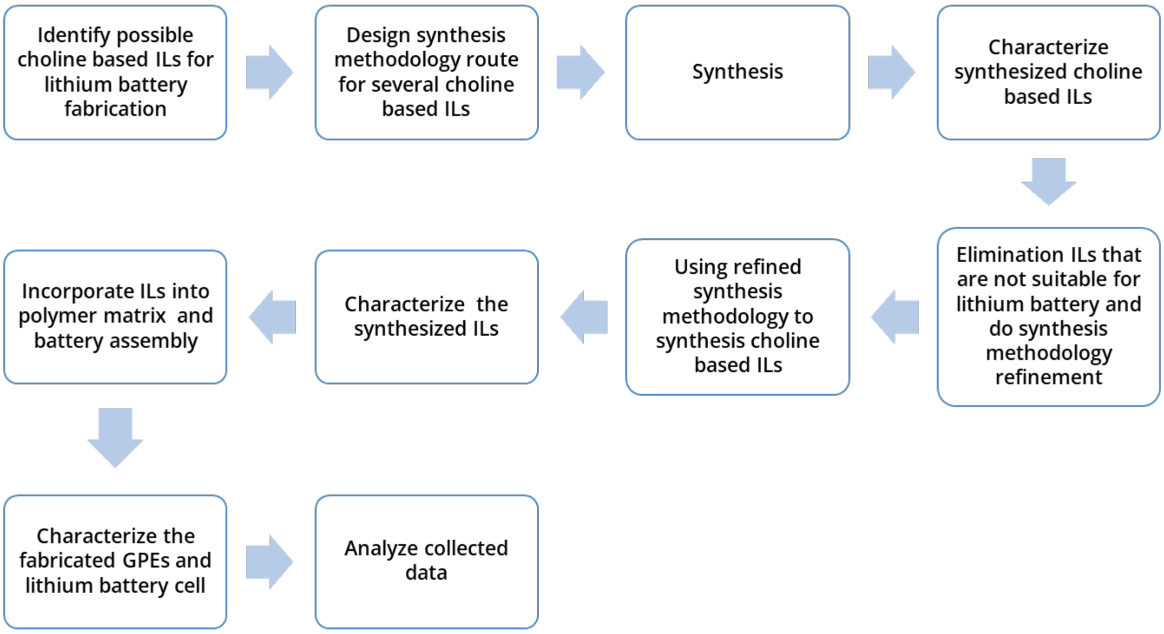
Key Novel Contributions:

1. Novel Choline-based IL Composition: This work explores new formulations of choline-based ILs with different anionic partners, which have not been widely studied in the context of GPEs for lithium-ion batteries. By fine-tuning the anion selection, the properties of the choline-based ILs can be tailored to achieve higher ionic conductivity and better compatibility with lithium-ion battery electrodes.

2. Integration with Gel Polymeric Matrix: While choline-based ILs are known, their incorporation into a gel polymeric matrix to form a stable electrolyte system is innovative. This study focuses on optimizing the interaction between the IL and the polymer matrix, improving the mechanical stability and ionic transport properties, which are crucial for practical applications in lithium-ion batteries.
3. Enhanced Safety and Environmental Sustainability: The focus on creating a safer, more environmentally friendly electrolyte is another key aspect of this work. The combination of choline's biodegradability and low toxicity with the structural benefits of a gel polymer matrix reduces the risks associated with leakage, flammability, and environmental damage, addressing safety concerns in conventional liquid electrolytes.
4. Improved Ionic Conductivity and Battery Performance: The novelty also lies in the improvement of the overall battery performance through enhanced ionic conductivity and electrochemical stability. By integrating choline-based ILs with a gel polymeric electrolyte, the research demonstrates improved lithium-ion transport, longer cycle life, and better thermal stability than traditional liquid electrolytes or other gel polymer systems.

In summary, while choline-based ILs have been explored in various contexts, the novelty of this research lies in the development of choline-based ionic liquids for gel polymeric electrolytes, focusing on innovative formulations, better interaction with polymer matrices, and enhanced safety and sustainability for lithium-ion battery applications.

1.7 Flow Diagram of the Research Methodology



1.8 Thesis Outline

Chapter 1: Introduction

- Background: Establish the urgent need for safer and more sustainable energy storage solutions, emphasizing the role of lithium ion batteries.
- Challenges with Current Electrolytes: Delineate the limitations of traditional liquid electrolytes, focusing on safety hazards (flammability, leakage) and potential performance constraints.
- Potential of Ionic Liquids and Polymer Electrolytes: Introduce ionic liquids as promising alternatives, highlighting their non-volatility, tunability, and potential for improved safety. Discuss how the incorporation of ionic liquids into polymer matrices, forming gel polymer electrolytes (GPEs), can further enhance safety and performance.

- Knowledge Gap: Identify the lack of systematic study on choline-based ionic liquids specifically for lithium battery electrolyte applications.
- Research Objectives and Approach: Clearly articulate the central research objectives, which should explicitly address the development of novel choline based ILs and GPEs and their optimization for battery use. Outline the overall strategy, including synthesis, characterization, and electrochemical evaluation.
- Thesis Statement: Present a concise and compelling thesis statement summarizing the overarching problem your research seeks to address and its significance.

Chapter 2: Literature Review

- Lithium ion Batteries: Provide a comprehensive overview of lithium ion battery technology, including working principles, components, and performance metrics. Discuss current challenges and limitations of lithium ion battery electrolytes.
- Ionic Liquids: Thoroughly examine the fundamentals of ionic liquids, their synthesis methods, and their physicochemical properties relevant to electrolyte performance. Survey existing choline-based ionic liquids, highlighting those with promising characteristics.
- Gel Polymer Electrolytes (GPEs): Explore the concept of GPEs, their advantages, and the different polymer matrices used. Discuss strategies for enhancing GPE performance, such as the use of fillers and additives, crosslinking, and fabrication methods.

Chapter 3: Experimental Methodology

- Materials: List all chemicals, reagents, and polymers used, including their specifications (purity, supplier, etc.).

- Synthesis of Ionic Liquids: Provide detailed step-by-step procedures for the synthesis of each choline-based IL, including reaction conditions, purification methods, and safety considerations.
- Fabrication of GPEs: Describe the methods for preparing GPEs, including polymer choice, solvent systems, and IL incorporation techniques.
- Characterization Techniques: Itemize the characterization methods employed, along with instrument models and specific settings. These techniques should encompass:
    - Structural analysis (NMR, FTIR,)
    - Thermophysical properties (TGA, DSC, viscosity)
    - Electrochemical properties (conductivity, electrochemical window, cycling tests)
    - Safety assessments (flammability)
    - Preliminary mechanical assessment

Chapter 4: Results and Discussion

- Synthesis and Structural Confirmation: Present results confirming successful IL synthesis (NMR, FTIR, etc.). Discuss any challenges encountered and modifications made to the synthesis procedures.
- Choline-based IL Properties: Analyze the thermophysical and electrochemical properties of the ILs. Critically evaluate their suitability for lithium battery electrolytes based on established benchmarks.
- Optimization of GPE Composition: Discuss the influence the alkyl chain length to the ionic conductivities. Identify the most promising GPE formulations.

- Performance Evaluation: Compare the key performance metrics of the optimized GPEs to both commercial and benchmark GPE systems reported in the literature.
- Structure-Property Relationships: Elucidate the connections between the molecular structure of the choline-based ILs, the GPE morphology, and their ultimate performance characteristics.

Chapter 5: Conclusion and Future Perspectives

- Summary of Findings: Concisely recap the major conclusions drawn from the research.
- Achievement of Objectives: Explicitly address whether the research objectives outlined in Chapter 1 were successfully met.
- Significance and Implications: Highlight the broader impact of your work on advancing lithium battery technology and the field of electrolyte design.
- Future Directions: Suggest pathways for further research, including potential optimization strategies, exploration of different choline-ester ILs, and investigations into long-term battery cycling performance.

Chapter 2: Literature Review

2.1 Advancement and Challenges in Lithium Ion Batteries (LIBs) Electrolyte

Electrolytes for lithium ion batteries (LIBs) can be categorized into several categories, namely organic electrolytes, aqueous electrolytes, gel polymer electrolytes, and solid-state electrolytes. Each type offers distinct advantages and challenges, contributing to numerous research on LIBs and their applications.

2.1.1 Organic Electrolyte

Organic electrolytes are essential components in LIBs, serving as the conductive media for ion transport between the cathode and anode. These electrolytes typically comprise organic carbonates, such as ethylene carbonate (EC) and dimethyl carbonate (DMC), dissolved with lithium salts to enable ionic conductivity (Cao, He and Kyu, 2017).

Common electrolyte solvents include a blend of cyclic carbonates (like EC and propylene carbonate (PC)) and linear carbonates (like DMC, diethylene carbonate (DEC), and ethyl methyl carbonate (EMC)). These carbonates exhibit low viscosities for suitable ionic conductivity (Lee *et al.*, 2020). EC is particularly valuable due to its high thermal stability, dielectric constant, and film-forming ability on the anode surface (P. Shi *et al.*, 2017). However, EC's high viscosity and melting point limit its ionic conductivity at room temperature, necessitating its use in combination with linear carbonates (Pohl and Wiemhöfer, 2015). Linear carbonates, despite their lower viscosities and dielectric constants, often create unstable solid electrolyte interface (SEI) layers, making them unsuitable for sole use with graphite anodes(S. Li *et al.*, 2018).

To improve LIB safety, researchers are exploring alternatives to traditional organic carbonates. A common strategy involves introducing electron-withdrawing groups like fluorine, sulfone, or cyano into organic molecules for enhanced stability against oxidation (Kalhoff *et* *al.*, 2015). Fluorinated carbonates, such as fluorinated ethylene carbonate (FEC), offer improved thermal stability, flame retardancy, and often contribute to a more robust SEI than their non-fluorinated counterparts (Gao *et al.*, 2020). However, their increased viscosity can hinder ionic conductivity (Olson *et al.*, 2016).

Lithium hexafluorophosphate ($LiPF_6$) is the predominant conductive salt in commercial LIBs. Its popularity stems from its wide electrochemical stability window, good ionic conductivity, ability to passivate aluminium current collectors, and low cost (Kalhoff *et al.*, 2015). However, $LiPF_6$ poses safety concerns due to its thermal instability, generation of highly reactive and toxic hydrogen fluoride (HF) when exposed to moisture, and its potential contribution to electrolyte decomposition at elevated temperatures (Min *et al.*, 2023). Research into alternative salts such as lithium bis(fluorosulfonyl)imide (LiFSI) focuses on mitigating these safety risks while maintaining suitable conductivity and stability (Heckmann *et al.*, 2016). While organic electrolytes remain vital components in LIBs, their inherent flammability, volatility, and potential for thermal instability are major safety concerns. Developing safer, more stable organic electrolytes while ensuring optimal electrochemical performance is an ongoing research focus. Continued advancements in novel solvents, electrolyte additives, and alternative lithium salts will be crucial to unlocking the full potential of LIBs for safe and widespread applications.

2.1.2 Aqueous Electrolyte

Aqueous lithium ion batteries (ALIBs) offer inherent safety advantages over traditional LIBs due to their non-flammable nature, making them attractive for various applications. In ALIBs, a concentrated saline solution serves as the electrolyte, facilitating lithium ion (Li-ion) transport between electrodes (Jeon and Cho, 2021). However, their electrochemical stability window (ESW) is limited (typically ~1.23V), leading to lower energy density compared to lithium ion batteries with organic electrolytes (Liu *et al.*, 2021).

To address the challenges of aqueous electrolytes, researchers developed water-in-salt (WIS) electrolytes. These highly concentrated solutions exhibit wider ESWs (up to 3V) and suppress water's chemical activity, improving battery stability (Becker, 2022). Lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) is a commonly used lithium salt in WIS electrolytes for its high reversibility, wide ESW, and the stability of its TFSI anion against hydrolysis and elevated temperatures (Khalid *et al.*, 2023). Other promising WIS electrolytes include lithium (pentafluoroethanesulfonyl)-(trifluoromethanesulfonyl)imide (LiPTFSI), lithium nitrate ($LiNO_3$), and lithium sulfate (Li_2SO_4) (Becker, Kühnel and Battaglia, 2019). WIS electrolytes can enhance electrochemical performance and stability when paired with specific anode and cathode materials. Incorporating additives and forming a stable solid electrolyte interface can further improve their stability, enabling their use with high-energy cathodes (Khalid *et al.*, 2023).

While aqueous electrolytes offer inherent safety and cost advantages, their limited voltage stability and susceptibility to electrode corrosion present challenges (Pan *et al.*, 2021). Continued research on WIS electrolytes, electrolyte additives, and protective coatings hold promise for improving the electrochemical performance and long-term stability of ALIBs, paving the way for their wider adoption in safe and sustainable energy storage solutions.

2.1.3 Gel Polymeric Electrolytes (GPEs)

Gel polymer electrolytes (GPEs) represent a significant advancement in battery technology, offering a balance between the conductivity advantages of liquid electrolytes and the structural integrity of solid electrolytes. At their core, GPEs consist of a polymer matrix that hosts a liquid electrolyte, typically comprising organic/inorganic salts dissolved in either water or organic solvents. This hybrid design results in several desirable properties, including reduced leakage risks during battery assembly and significantly higher ionic conductivities (10^{-4}–10^{-3} S cm^{-1}) compared to purely solid electrolytes (Li, Qiao and Lian, 2020; Liu, Borodin and Endres, 2021). The versatility of GPEs stems from the diverse selection of polymers that can serve as the matrix.

Ether-based polymers, such as poly(ethylene oxide) (PEO), are a popular choice due to their environmental friendliness and cost-effectiveness (Herzberger *et al.*, 2016). However, PEO tends to exhibit high crystallinity, which limits ionic conductivity at room temperature (Wei *et al.*, 2018). Researchers combat this by introducing inorganic fillers (like SiO_2, TiO_2, LLZO) to disrupt crystallinity or by employing grafting and crosslinking strategies to modify the molecular structure of the polymer (K. Luo *et al.*, 2021). Another class of polymers, ester-based polymers like poly(methyl methacrylate) (PMMA), are generally inexpensive but suffer from brittleness. They often require copolymerization with other polymers to improve their mechanical properties (Riazi, Mohammadi and Mohammadi, 2013).

Nitrile-based polymers, exemplified by polyacrylonitrile (PAN), boast exceptional mechanical strength and a wide electrochemical stability window (exceeding 4.5 V vs. Li+/Li), making them promising for high-voltage applications (Hu *et al.*, 2016). The drawback of PAN, however, is its poor compatibility with lithium metal, limiting its direct use in lithium metal batteries (Hu *et al.*, 2016). Blending PAN with inorganic fillers like TiO_2 can enhance its ionic conductivity (Kumar, Sahu and Mahipal, 2023). Fluorine-based polymers, primarily poly(vinylidene fluoride) (PVDF) and its copolymer Poly(vinylidene fluoride-co-hexafluoropropylene) PVDF-HFP, offer excellent electrochemical stability and toughness (Barbosa *et al.*, 2018). Their crystallinity can be reduced by the addition of amorphous HFP or inorganic fillers, leading to improvements in conductivity (Barbosa *et al.*, 2022).

Beyond these common substrates, researchers explore hydrogels based on polyvinyl alcohol (PVA), polyacrylic acid (PAA), and polyacrylamide (PAM) for applications in flexible supercapacitors (Y. Li *et al.*, 2022). PVA is known for its non-toxicity, biocompatibility, and good mechanical strength, while PAA and PAM offer enhanced hydrophilicity for better cation transport (Cheng *et al.*, 2021). The choice of plasticizer within GPEs is another crucial factor(J. Zheng *et al.*, 2023). Organic solvents improve both GPE conductivity and the operating voltage of energy storage devices (Bhat and Hashmi, 2022). Ionic liquids (ILs) excel in polymer compatibility and reduce plasticizer evaporation, leading to more stable performance (Z. Wang *et al.*, 2020). The ongoing development of GPEs, with its focus on polymer design, innovative plasticizers, and the integration of functional fillers, promises to unlock safer, higher-energy-density, and more flexible batteries for a wide range of applications.

2.1.4 Solid State Electrolyte

Solid-state electrolytes (SSEs) are the driving force behind the development of safer and more powerful solid-state batteries. Unlike flammable and volatile liquid electrolytes, SSEs offer enhanced safety and the potential for higher energy densities (Yao *et al.*, 2020). For instance, they allow the use of lithium metal anodes, which boast a significantly higher theoretical capacity (3861 mAh g^{-1}) compared to traditional graphite (Qian *et al.*, 2022). Additionally, SSEs exhibit a wider operating temperature range and the potential to suppress hazardous lithium dendrite growth (Cao *et al.*, 2020). However, challenges remain. While sulfide-based SSEs boast exceptional ionic conductivities (sometimes exceeding liquid electrolytes), they require careful handling due to moisture sensitivity (C. Wang *et al.*, 2021). Oxide-based SSEs, known for their chemical stability, include several families: oxysalts like LISICON (e.g., $Li_{14}Zn(GeO_4)_4$) and LiPON (used in thin-film batteries), NASICON-type compounds (e.g., $Li_{1.3}Al_{0.3}Ti_{1.7}(PO_4)_3$ with $\sim 10^{-3}$ S cm^{-1} conductivity), perovskites (such as $Li_{3x}La_{2/3-x}TiO_3$), and garnet-types like $Li_7La_3Zr_2O_{12}$ (stable against lithium metal) (Zhai *et al.*, 2022).

Researchers are tackling challenges for the limited SSE ionic conductivity and high interfacial resistance between SSEs and electrodes (Tao *et al.*, 2022). Strategies include creating composite structures where oxide SSEs are combined with lithium oxysalts (like Li_3BO_3) to reduce grain boundary resistance (Cretu *et al.*, 2023). Interfacial engineering, such as introducing thin layers (e.g., Al_2O_3) and using precycling techniques, can improve compatibility with lithium metal anodes (Zhang *et al.*, 2023). Lastly, the field continues to explore novel materials, dopant substitutions, and innovative synthesis methods to optimize the bulk conductivity of SSEs and minimize defects contributing to grain boundary resistance (Feng *et al.*, 2022).

The future of SSEs is bright, and continued advancements promise to unlock safer, more energy-dense, and longer-lasting batteries with transformative potential for electric vehicles and grid-scale energy storage.

2.1.5 Solid Electrolyte Interphase (SEI) in LIBs

In lithium ion batteries, the longevity, safety, and performance are critically dependent on the formation and stability of a thin film known as the solid electrolyte interphase (SEI) (Shan *et al.*, 2021). This passivating layer forms spontaneously on the surface of reactive electrode materials, particularly the anode. It arises from the electrochemical reduction of electrolyte components and serves as a crucial barrier between the electrode and the electrolyte (A. Wang *et al.*, 2018). Understanding the SEI is essential for designing advanced lithium ion batteries with improved durability, power density, and safety (Adenusi *et al.*, 2023). The ideal SEI strikes a delicate balance between several competing requirements. It must be an excellent electronic insulator to prevent continuous electrolyte decomposition and parasitic reactions that consume active lithium and reduce battery capacity (Quilty *et al.*, 2023). Simultaneously, the SEI must be a good ionic conductor, selectively allowing the passage of lithium ions to facilitate the charge and discharge processes (Lee *et al.*, 2023). To withstand the volume changes that electrodes experience during cycling, a robust SEI possesses both chemical and mechanical stability. This stability helps avoid continuous SEI reformation, which can irreversibly deplete the electrolyte (Jiang *et al.*, 2020). A thin and uniform SEI layer is also beneficial, as it minimizes interfacial resistance and promotes efficient ion transport (Meda *et al.*, 2022).

The SEI forms immediately upon the first contact between the electrode and electrolyte and continues to evolve throughout the initial cycling of the battery (An *et al.*, 2016). Its complex composition is influenced by a multitude of factors. The specific electrolyte chemistry, including the solvent(s), lithium salt(s), and any additives, plays a major role in determining the SEI components (Adenusi *et al.*, 2023). For example, the use of ethylene carbonate (EC) as a solvent often leads to the formation of lithium ethylene dicarbonate (LEDC) within the SEI (Xing *et al.*, 2018). The electrode material's surface chemistry and reactivity further dictate the nature of the reduction reactions that form the SEI (An *et al.*, 2016). Additionally, operating conditions such as temperature, cycling rate, and the depth of charge and discharge also influence SEI stability and growth (Z. Liu *et al.*, 2016). The SEI typically consists of a mosaic of inorganic components, such as LiF, Li_2CO_3, and Li_2O, interspersed with organic species like lithium alkyl carbonates and polymeric compounds (Haas *et al.*, 2022).

Despite its protective role, the SEI is prone to failure, leading to several detrimental consequences for battery performance and safety. Elevated temperatures can lead to the decomposition of SEI components, resulting in a non-uniform, porous layer that does not effectively passivate the electrode (M. Li *et al.*, 2023). Mechanical stresses arising from volume changes during cycling can cause cracking and fracturing of the SEI (Deshpande and Bernardi, 2017). This exposes fresh electrode surfaces to the electrolyte, triggering further SEI formation and the consumption of both active lithium and electrolyte components, ultimately leading to capacity fade and decreased battery life. One of the most concerning SEI-related issues is the formation of lithium dendrites. These needle-like metallic protrusions can grow through imperfections or instabilities in the SEI (B. Li *et al.*, 2023).

2.1.6 Thermal Runaway in LIBs

The remarkable progress of lithium ion batteries (LIBs) has fuelled their widespread adoption in portable electronics, electric vehicles, and energy storage applications. However, the increasing incidence of LIB-related fires and explosions poses a significant barrier to their further implementation. A critical factor in these incidents is thermal runaway, a cascade of exothermic reactions within the battery that can lead to catastrophic failure. Recent research highlights the pivotal role of liquid electrolyte chemistry (LEC) in both initiating and exacerbating thermal runaway events (Chavan *et al.*, 2023).

Thermal runaway begins with the degradation of the solid electrolyte interface (SEI) and reactions between the electrolyte and lithiated anode, occurring at temperatures as low as 60-80 ºC (Allcorn and Manthiram, 2015). These reactions release reductive gases such as hydrogen (H_2), ethene (C_2H_4), and propene (C_3H_6) which can migrate to the cathode and compromise its thermal stability (Yu Wang *et al.*, 2023). This cross talk between the electrodes leads to primary heat accumulation within the cell. As temperatures rise, the separator may collapse, leading to an internal short circuit (Huang *et al.*, 2021). Subsequently, the flammable liquid electrolyte evaporates, and the cathode material decomposes, potentially releasing highly reactive oxygen (O_2) (Zhang, Li and Lu, 2021). These exothermic reactions further escalate the temperature, ultimately resulting in the venting of flammable gases, fire, and potential explosion.

To address thermal runaway risks, researchers are focusing on developing functional liquid electrolytes (LEs) that target the specific reactions contributing to this hazardous event. Key strategies include:

- Promoting Thermally Stable SEI: A breakdown of the SEI layer and its subsequent reconfiguration can release reductive gases. LEs that promote a robust and thermally stable SEI are crucial for minimizing gas generation, reducing cross talk between electrodes, and preventing the initial heat buildup. Strategies to achieve this include using additives that undergo polymerization reactions at elevated temperatures. For example, methyl difluoroacetate (MFA) (Zu, Yu and Li, 2021)or 1,3,5-trimethyl-1,3,5-tris(3,3,3-trifluoro propyl)-cyclotrisiloxane (D_3F) (M. Liu *et al.*, 2023) can form highly stable polymerized layers that protect the electrodes.

- Utilizing Non-Flammable Electrolytes: Conventional carbonate-based electrolytes are highly flammable, posing significant risks for secondary disasters involving combustion and explosion. Non-flammable electrolytes address this issue by eliminating both fuel and oxygen radicals. Promising alternatives include phosphate-based electrolytes, fluorinated solvents, and highly concentrated electrolytes, although each has its own challenges. Phosphate-based electrolytes offer excellent flame-retardant properties but often suffer from poor film-forming ability and compatibility issues within the cell (Gond *et al.*, 2021). While fluorinated solvents and highly concentrated electrolytes can enhance safety, their high cost and potential for reduced conductivity remain hurdles to commercialization (J. Liu *et al.*, 2023).
- Establishing Robust Cathode Electrolyte Interface (CEI): The release of reactive oxygen (O_2) from a deteriorating cathode structure is a major contributor to thermal runaway. Advanced LEs designed to form a strong and effective CEI are essential. This CEI acts as a protective barrier, shielding the cathode material from direct contact with the electrolyte and preventing irreversible phase transitions that lead to oxygen release (Liu *et al.*, 2021). The use of high-voltage electrolyte systems (beyond 4.4 V) can facilitate the formation of robust CEIs capable of withstanding the harsh electrochemical environment at the cathode (M. Liu *et al.*, 2023).

Despite promising advances, challenges remain in developing commercially viable functional LEs. Low-cost polymeric solvents for SEI stabilization are needed. Additionally, the trade-offs between safety, conductivity, and compatibility in non-flammable electrolyte systems must be carefully balanced. While high-voltage electrolytes improve CEI stability, their inherent reactivity and limited oxidative stability require further optimization.

The ideal future electrolyte would combine these functionalities to offer comprehensive protection against thermal runaway. Continued research focusing on the intricate relationships between LEC and thermal runaway mechanisms, along with the development of novel theoretical models and electrolyte design concepts, holds the key to unlocking the full potential of LIBs. Critical research areas include investigating the applicability of functional LEs in larger-capacity batteries, assessing their long-term performance in full cells, and developing integrated safety mechanisms that can provide early warnings of potential thermal runaway events.

2.1.7 Summary

Lithium ion battery (LIB) electrolytes are crucial for enabling ion transport between the electrodes. Current research focuses on improving their safety, stability, and performance. Organic electrolytes, while widely used, are inherently flammable. Researchers are exploring safer alternatives, like fluorinated carbonates that improve thermal stability and form protective surface layers. Aqueous electrolytes provide intrinsic safety but are limited by their narrow voltage window. Concentrated 'water-in-salt' electrolytes expand this window and improve their compatibility with diverse electrode materials.

Gel polymer electrolytes (GPEs) offer a compromise between solid and liquid electrolytes, combining good conductivity with reduced leakage risks. The choice of polymer matrix (e.g., PEO, PMMA, PAN, PVDF) influences conductivity, stability, and compatibility with various electrode chemistries. Solid-state electrolytes (SSEs) promise exceptional safety and the potential for high energy density, but overcoming limitations of ionic conductivity and interfacial resistance is an active research area.

Finally, understanding the solid electrolyte interphase (SEI) and managing thermal runaway are vital for safe and long-lasting LIBs. Strategies for SEI control involve electrolyte additives and engineering the electrode-electrolyte interfaces. Addressing thermal runaway focuses on developing functional liquid electrolytes that promote stable SEI formation, utilize non-flammable components, and create robust barriers at the cathode (CEI).

2.2 Advancement and Challenges in Ionic Liquids (ILs) as Electrolytes

2.2.1 Ionic Liquids (ILs)

Lithium ion batteries (LIBs) are essential for modern portable electronics and the burgeoning electric vehicle industry. However, traditional LIB electrolytes, composed of lithium hexafluorophosphate ($LiPF_6$) in carbonate solvents, suffer from safety hazards due to their flammability and poor thermal stability (He *et al.*, 2019). Moreover, their relatively narrow electrochemical windows limit the use of high-energy-density materials (D. Wang *et al.*, 2020). ILs offer a promising alternative electrolyte solution due to their non-flammability, low vapor pressure, good electrochemical stability, and wide electrochemical windows (Sivaji, Vijayalakshmi and George, 2019).

ILs are molten salts with melting points below 100 °C, with many exhibiting liquid states at room temperature(Guan *et al.*, 2019; Yong-Lei Wang *et al.*, 2019; Gutiérrez-Serpa *et al.*, 2020). They typically consist of bulky, asymmetric organic cations (e.g., imidazolium, pyrrolidinium, piperidinium, and quaternary ammonium) paired with inorganic or organic anions (e.g., PF_6^-, BF_4^-, $TFSI^-$, FSI^-) (Barbosa, Costa and Lanceros-Méndez, 2019; Correia *et al.*, 2019). The unique chemical structures of ILs strongly influence their physicochemical properties and, in turn, their performance in LIBs.

2.2.1.1 Structure and Properties of IL-Based Electrolytes

The cationic component in ILs significantly affects properties like cathodic stability and lithium diffusion (Kim *et al.*, 2017). Imidazolium-based ILs, particularly those with short alkyl chains, generally exhibit lower viscosity and higher ionic conductivity than other ILs (Khalil *et al.*, 2020). For example, the conductivity of 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide (EMIM-TFSI) can reach around 10 mS cm^{-1} (Yang *et al.*, 2020). However, their electrochemical stability is limited to approximately 4 V vs. Li/Li^+, which hinders their use with high-voltage cathode materials. In contrast, quaternary ammonium ILs possess wider electrochemical stability windows (≥5 V vs. Li/Li+), but often suffer from poorer ionic conductivity due to their larger size (Kazemiabnavi *et al.*, 2016).

Anions also play a critical role. Highly fluorinated anions, such as PF_6^- and BF_4^-, tend to be more electrochemically stable but can generate corrosive hydrogen fluoride (HF) upon hydrolysis (Aangothu *et al.*, 2019; C. Wang *et al.*, 2022). Imide-type anions, such as $TFSI^-$ and FSI^-, exhibit good electrochemical stability, relatively low viscosity, and high conformational flexibility, making them well-suited for battery electrolytes (Oteo *et al.*, 2019).

The size and structure of anions affect ion transport in IL-based electrolytes. FSI^- anions, with a smaller radius (~0.264 nm) than $TFSI^-$ (~0.379 nm), lead to electrolytes with lower viscosities and higher ionic conductivities. Importantly, the coordination between Li^+ cations and anions influences charge transfer at the electrode-electrolyte interface (K. Liu *et al.*, 2021a).

2.2.1.2 Physiochemical Properties of IL-Based Electrolytes

ILs exhibit superior thermal stability compared to conventional carbonate solvents, with onset decomposition temperatures (T_{onset}) often exceeding 200 °C (Tang *et al.*, 2022). For instance, 1-butyl-3-methylimidazolium TFSI (BMIM-TFSI) remains stable up to 430 °C (Shalu, Singh and Singh, 2015; V. Singh *et al.*, 2018). This enhanced thermal stability significantly improves battery safety.

A key drawback of ILs is their relatively high viscosity stemming from their large organic cations. For example, *n*-methyl-*n*-propylpyrrolidinium bis(trifluoromethanesulfonyl)imide ($PYR_{13}TFSI$) has a room temperature viscosity of approximately 60 mPa s (Plylahan *et al.*, 2016). This high viscosity impedes ion mobility and limits battery performance. However, IL-based electrolytes with viscosities as low as 20–60 cP have been formulated using ILs with dicyanamide, $TFSI^-$, or BF_4^- anions (Bhattacharjee *et al.*, 2014; Y. Zhang *et al.*, 2020).

2.2.1.3 Ionic Liquids as Electrolytes in Lithium Batteries

Research into IL-based electrolytes focuses on systems where lithium salts are dissolved in neat ILs. These binary electrolytes offer the advantages of ILs while ensuring the presence of Li^+ for battery operation. Their electrochemical stability windows are often wider (> 5V vs. Li/Li^+) than traditional electrolytes, enabling compatibility with high-potential electrodes (Cherkashinin *et al.*, 2018). For instance, ILs based on $PYR_{13}TFSI$ and lithium bis(trifluoromethylsulfonyl)imide (LiTFSI) show promise wide anodic limit of 5.8- 6 V for high-temperature (80 °C) LIBs (Rogstad, Einarsrud and Svensson, 2021).

Further advancements have explored dicationic ILs, which contain two cationic moieties linked by a hydrocarbon chain (Pagano *et al.*, 2012). Dicationic ILs often exhibit wider electrochemical stability windows, higher thermal degradation temperatures, and greater tunability compared to their monocationic counterparts (Lall-Ramnarine *et al.*, 2014, 2016).

2.2.1.4 Monocationic vs. Dicationic Ionic Liquids

The vast majority of research on IL-based electrolytes has centered on monocationic ILs (Chakraborty, Ahmed and Sarkar, 2019). These ILs contain a single cationic moiety paired with an anion. Common monocationic cations include:

- Imidazolium: Exhibiting relatively low viscosities and high conductivities (e.g., 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide (EMIM-TFSI) with conductivity around 10 mS cm^{-1}) but typically have electrochemical stability windows limited to around 4 V vs. Li/Li^+ (Yang *et al.*, 2020).
- Pyrrolidinium and Piperidinium: Offer wider electrochemical stability windows (≥5 V vs. Li/Li+) enabling compatibility with high-voltage cathodes. However, they often suffer from higher viscosities, which can hinder ionic conductivity (Sivaji, Vijayalakshmi and George, 2019; Lv *et al.*, 2020).
- Quaternary Ammonium: Similar to pyrrolidinium and piperidinium cations, quaternary ammonium ILs display desirable electrochemical stability but can have even greater viscosities that limit their practical application (Griffin *et al.*, 2014; Tian *et al.*, 2020).

The physicochemical and electrochemical properties of monocationic ILs can be fine-tuned through modifications to the cation (e.g., introducing electron-donating substituents or altering alkyl chain lengths) and by pairing different anions. Anions like $TFSI^-$ and FSI^- offer a balance of good conductivity, electrochemical stability, and relatively low viscosity (Kerner *et al.*, 2015; Montalbán *et al.*, 2015; Roohi and Ghauri, 2016).

Dicationic ILs, featuring two cationic moieties joined by a hydrocarbon linker, represent a more recent and less explored avenue in electrolyte design. These ILs offer several potential advantages:

- Enhanced Electrochemical Stability: Dicationic ILs can exhibit even wider electrochemical stability windows than quaternary ammonium-based monocationic ILs, with some up to 6 V vs. Li/Li^+. This allows for the exploration of higher voltage electrode materials and increased energy density (Lall-Ramnarine *et al.*, 2017; Vélez *et al.*, 2019).

- Tailored Properties: By altering the length of the alkyl linker, the substituents on the cationic moieties, and the choice of anion, dicationic ILs offer greater tunability compared to monocationic ILs. This flexibility can be used to optimize properties such as viscosity, conductivity, and thermal stability (Patil *et al.*, 2016; Chakraborty, Ahmed and Sarkar, 2019; Piper *et al.*, 2022).

- Asymmetrical Designs: Asymmetrical dicationic ILs combine different cation types (e.g., imidazolium and trialkylammonium). This strategy helps balance properties like conductivity and electrochemical stability – a valuable asset for electrolyte optimization (Masri, Mi and Leveque, 2016; Vélez *et al.*, 2018).

2.2.1.5 IL-Organic Solvent Hybrid Electrolytes

Hybrid electrolytes combine ILs with traditional volatile organic solvents to achieve a balance of properties and address the limitations of pure IL electrolytes. Common organic solvents used in these hybrids include:

- Carbonates: These include propylene carbonate (PC), ethylene carbonate (EC), dimethyl carbonate (DMC), and ethyl methyl carbonate (EMC) (Plylahan *et al.*, 2016; Wagaye, Yohannes and Workneh, 2023).

- Ethers: 1,3-dioxolane (DOL), 1,2-dimethoxyethane (DME), and tetrahydrofuran (THF) are examples (Angulakshmi *et al.*, 2021; Garcia-Quintana *et al.*, 2022).

- Other Solvents: Acetonitrile (ACN), sulfolane, and others may also be used (Décoppet *et al.*, 2014; Neale *et al.*, 2017; Osti *et al.*, 2023).

Hybrid electrolytes provide several benefits:

- Reduced Viscosity and Enhanced Conductivity: The inclusion of organic solvents significantly reduces electrolyte viscosity, promoting faster ion movement and improving battery performance, especially at low temperatures (Plylahan *et al.*, 2016; Phung *et al.*, 2021).
- Optimized SEI Formation: The synergistic interaction between ILs and organic solvents can lead to the formation of a robust SEI layer that protects the electrodes and enhances their stability (N. Li *et al.*, 2017; T. H. Wang *et al.*, 2022).
- Cost Reduction: Partially replacing ILs with less expensive organic solvents helps control electrolyte production costs.

Examples of commonly used IL-organic solvent hybrids include $PYR_{14}TFSI$ + EC/DMC mixtures offering balanced viscosity, conductivity, and electrochemical stability (Tartaj *et al.*, 2016; Kong *et al.*, 2018). Combining ILs with ethers like DOL or DME can create electrolytes with high anodic stability against oxidation (He *et al.*, 2022). However, incorporating volatile and potentially flammable organic solvents compromises the overall safety and thermal stability of the hybrid compared to a pure IL electrolyte. The choice of organic solvent and the IL-to-solvent ratio significantly influence the properties of hybrid electrolytes, necessitating careful selection to optimize performance characteristics.

2.2.1.6 IL-Water Hybrid Electrolytes

While less prevalent, IL-water hybrid electrolytes have gained interest for specific battery chemistries (Dhattarwal and Kashyap, 2022). These electrolytes incorporate a controlled amount of water, taking advantage of the hygroscopicity of ILs (Tang *et al.*, 2022).

- Reduced Viscosity and Enhanced Conductivity**:** Water molecules can alter the ion pairing within ILs, leading to reduced viscosity and improved ionic conductivity (Xu *et al.*, 2018).
- Modified SEI Composition**:** The presence of water can modify the composition and properties of the SEI formed on electrode surfaces (Dou *et al.*, 2020).
- Widened Electrochemical Window (in Specific Cases): Some IL-water systems, like the 'water-in-salt ionic liquid' (WiSIL) electrolytes, exhibit wider electrochemical stability windows than typical aqueous electrolytes (Zhang *et al.*, 2018).

An example of an IL-water hybrid electrolyte is the LiTFSI/water/EMIMTFSI system (WiSIL) with an electrochemical stability window of up to ~3.8 V (Dou *et al.*, 2020). This enhanced stability window makes it compatible with certain metal oxide anode materials.

2.2.2 Deep Eutectic Solvents (DESs)

DESs, a specific class of ionic liquids first developed by Abbott and co-workers, are sometimes considered a subclass of ionic liquids by certain researchers (Ju *et al.*, 2012; Reuter *et al.*, 2019; Skowrońska and Wilpiszewska, 2022). DESs are mixtures of at least two components that exhibit a significantly lower melting point than their individual constituents. This unique property arises from strong intermolecular interactions, including hydrogen bonding, Lewis acid-base interactions, and van der Waals forces (Jaumaux *et al.*, 2020). Typically, DESs contain a hydrogen bond donor (HBD), such as an amide or alcohol, and a hydrogen bond acceptor (HBA), like a metal halide or quaternary ammonium salt (Jia and Michinobu, 2023). The specific combination of components, their interactions, and their molar ratios make DESs highly tailorable, earning them the designation "designer solvents" (Cui *et al.*, 2019).

The physicochemical and electrochemical properties of DESs make them attractive for electrolytes in batteries and other energy storage devices. By carefully selecting HBDs and HBAs, researchers can tune properties like conductivity, viscosity, electrochemical stability, and environmental compatibility. These systems often exhibit low volatility, high thermal stability, and a wider electrochemical window than traditional organic electrolytes, offering potential safety and performance advantages (Miller, Wainright and Savinell, 2017).

The versatility of DESs extends beyond their tunability as electrolytes. Due to their advantageous properties, they are gaining interest in various fields. Researchers are exploring DESs as "green" solvents for synthesis, catalysis, and the recovery of materials from spent batteries (Ndizeye, Suriyanarayanan and Nicholls, 2019; Suriyanarayanan *et al.*, 2023). Their unique ability to dissolve a broad range of substances makes them valuable tools in both industrial and energy applications.

The physicochemical properties of DESs are of particular importance for their electrolyte applications. Factors like density, viscosity, surface tension, and ionic conductivity directly impact performance in energy storage devices (Wu *et al.*, 2021). The electrochemical stability window (ESW) determines the range of voltages at which the electrolyte remains stable, influencing its compatibility with different electrode materials. Additionally, environmental considerations like the biodegradability and toxicity of DES components are increasingly important for sustainable energy solutions (Ge *et al.*, 2017; Atilhan and Aparicio, 2021).

DESs bring several key advantages to LIBs. Firstly, their non-flammability and low volatility significantly reduce the risk of fires and explosions, enhancing battery safety (Wu *et al.*, 2021). This improvement makes them particularly attractive for applications where safety is critical, such as electric vehicles or large-scale energy storage systems. Secondly, DESs often demonstrate a wide electrochemical window (EW). For example, electrolytes based on LiTFSI/MAc (N-methylacetamide) have an EW of 4.7V vs. Li^+/Li, allowing the use of higher voltage cathode materials that can further increase a battery's energy density (Puttaswamy *et al.*, 2022). Additionally, DESs typically exhibit good thermal stability, allowing for operation across a wider range of temperatures without compromising performance (Wu *et al.*, 2021). Finally, the components used in DESs are often readily available and relatively inexpensive, offering the potential for cost-effective electrolyte solutions compared to traditional counterparts (Chakrabarti *et al.*, 2014; Hariyanto *et al.*, 2023).

Numerous studies have demonstrated how DESs can improve the performance of LIBs. DESs can achieve ionic conductivities suitable for many applications. For instance, LiTFSI/MAc DES electrolyte offers conductivity in the range of 0.76 - 1.41 $mScm^{-1}$, particularly at elevated temperatures (Zaïdi *et al.*, 2014). When used in batteries, DES formulations have shown impressive results. A battery utilizing LiTFSI/MAc electrolyte with a lithium iron phosphate (LFP) cathode and lithium titanate (LTO) anode achieved a discharge capacity of approximately 100 $mAhg^{-1}$ at 1 °C (Boisset *et al.*, 2013). Researchers have developed DES-based GPEs, where the DES is integrated into a polymer host. These GPEs, with optimized compositions like 74% polyethylene glycol methyl ether acrylate (MPEGA), 24% 2-hydroxyethyl acrylate (HEA), and 2% poly (ethylene glycol) diacrylate (PEGDA), have exhibited improved discharge capacity in LTO | | LMO full cells and excellent cycling stability with capacity retention above 75% at 0.45 C (Logan *et al.*, 2020).

Researchers are continuously exploring strategies to further enhance the performance of DES-based electrolytes. One approach involves developing solid composite electrolytes (SCEs), such as those combining DESs with silica matrices. ETGs (Eutecto Gels), incorporating a LiTFSI:MAc DES within a silica framework, exhibit ionic conductivities between 0.07 – 1.46 $mScm^{-1}$ at 25 °C and electrochemical windows as wide as 1.1 – 4.7 V vs.

Li^+/Li (Joos *et al.*, 2018). When applied in a lithium half-cell with an LFP cathode, these ETGs enable stable cycling performance with capacities exceeding 100 $mAhg^{-1}$. Similarly, polymetric eutectogels (P-ETGs), using a DES encapsulated within a polymer backbone, have achieved conductivities of 0.029 – 0.76 $mScm^{-1}$ at room temperature (Joos *et al.*, 2020).

DESs offer a promising route towards non-flammable electrolytes for lithium metal batteries (LMBs). However, their practical use is hindered by degradation when in contact with lithium metal at low voltages (W. Li *et al.*, 2022). Here, W. Li et al have demonstrated a strategy to improve DES stability and compatibility with lithium metal anodes. By utilizing fluoroethylene carbonate (FEC), a film-forming compound, as a cosolvent in DES-based electrolytes, a robust and compact solid electrolyte interphase (SEI) layer forms on the anode surface. The optimal electrolyte formulation, termed “50%DES”, retains a high degree of flame retardancy and exhibits a Li^+ conductivity of 1.58 mS cm^{-1} at room temperature. Importantly, introducing 50% FEC drastically reduces the viscosity of the DES electrolyte from 1206.3 mPa s^{-1} to a mere 20.4 mPa s^{-1}. This improved fluidity benefits ion transport within the battery system. When tested in a $Li/LiFePO_4$ cell, the 50%DES electrolyte demonstrated a reversible discharge capacity of 140.7 mAh g^{-1} at 50 mA g^{-1} with a capacity retention of 94.3% after 200 cycles. This impressive cycling stability results from the robust SEI layer formed on the lithium metal anode. This layer is rich in inorganic compounds such as LiF and Li_3N, which facilitate rapid Li^+ transport, improve interfacial stability, and crucially, suppress the formation of lithium dendrites. This work highlights the potential of DES and FEC mixtures as nonflammable electrolytes for safer, high-performance LMBs (W. Li *et al.*, 2023).

Further advancements in DES-based electrolytes are driven by in-depth investigations into their unique properties and behaviors. Srinivasan et al. delve into the mechanisms governing lithium ion solvation and transport within a DES composed of lithium perchlorate and acetamide. Using molecular dynamics (MD) simulations and validated by neutron scattering, they revealed two dominant states of acetamide in the DES:

1. Lithium-bound acetamide**:** Approximately 36% of acetamide molecules are hydrogen-bonded (H-bonded) to lithium ions. These exhibit significantly slower long-range diffusion compared to free acetamide molecules.
2. Free acetamide**:** Roughly 58% of acetamide molecules exist without H-bonds and move more freely.

Lithium ion dynamics in this DES are strongly influenced by acetamide. On average, each lithium ion is H-bonded to around 3.2 acetamide molecules within its first solvation shell. Importantly, analysis of H-bond correlation functions reveals that approximately 90% of lithium ion transport occurs via vehicular motion; the ions diffuse along with their bound solvation shells. Only about 10% of lithium ion transport is attributed to structural diffusion, where ions move by exchanging molecules within their solvation shells (Srinivasan *et al.*, 2020).

Unraveling these mechanisms is critical, as ionic conductivity and correlations directly impact the performance of DES-based electrolytes. MD simulations indicate that ions in this DES are largely uncorrelated, suggesting dissociation between anions and cations. The calculated ionic conductivity is 11 $mScm^{-1}$ (Srinivasan *et al.*, 2020).

These findings highlight the interplay between acetamide states and lithium ion transport. The insights suggest pathways for improving DES performance, such as:

- Tuning the DES: Modifying the hydrogen bond donor (HBD), anion, or adding water could influence viscosity and improve ion mobility.

- Polymer networks: Introducing polar polymer networks might enhance structural diffusion, boosting overall conductivity.

The future of DESs in lithium ion batteries is promising. Their potential to address the key safety concerns of conventional LIBs, combined with their thermal stability and potential cost-effectiveness, make them exciting avenues for research and development. Ongoing efforts to discover novel DES formulations, implement creative optimization strategies, and integrate DESs into advanced battery architectures are crucial steps toward unlocking the full potential of DESs and enabling a new generation of safer and higher-performance lithium ion batteries.

2.2.3 Polymeric ionic liquids (PILs)

Polymeric ionic liquids (PILs), a unique class of polymers formed by polymerizing ionic liquid (IL) monomers, offer a promising alternative (Q. X. Shi *et al.*, 2017). PILs inherit the favorable properties of ILs, such as non-flammability, thermal stability, and electrochemical stability (Suo *et al.*, 2021). Importantly, their polymeric form grants them solid-state characteristics, film-forming ability, tunable properties, and processability, overcoming the limitations of liquid IL electrolytes (Correia *et al.*, 2020).

Researchers have extensively explored PIL-based gel polymer electrolytes, where PILs are combined with IL-based electrolytes (a lithium salt dissolved in an IL). These GPEs exhibit enhanced ionic conductivity, essential for LIB performance. The chemical affinity between PILs and ILs prevents phase separation within the GPE, ensuring homogenous properties and mitigating electrolyte leakage. For example, Nykaza et al. have synthesized a novel polymerized ionic liquid diblock copolymer (PILBCP-TFSI) consisting of ionic and non-ionic monomer blocks. This material can form mechanically stable films that, when combined with lithium salt and an ionic liquid (Li-TFSI/EMIm-TFSI), act as a solid-state electrolyte (SSE). These films achieve high ionic conductivities (>1 mS cm^{-1} at room temperature), exceeding the performance of many liquid electrolytes. Importantly, this PILBCP-TFSI electrolyte demonstrates its functionality as both electrolyte and separator in lithium ion batteries. Test batteries assembled with this SSE exhibited a maximum discharge capacity of 112 mAh g^{-1} at room temperature. The batteries also maintained a Coulombic efficiency above 94% over 100 charge-discharge cycles. These results highlight the feasibility of using PIL diblock copolymers as solid-state electrolytes in lithium ion batteries. While the current electrochemical stability window (2.7 V) limits long-term performance, this work demonstrates the potential of PIL-based SSEs. With further optimization to enhance stability, these materials could open avenues for safer and more efficient battery designs (Nykaza *et al.*, 2016).

To further optimize PIL-based electrolytes, several strategies are employed. In-situ plasticization, such as the copolymerization of the IL monomer 1-vinyl-3-butylimidazolium bis-(trifluoromethylsulfonyl)imide with poly(ethylene glycol) diacrylate, reduces crystalline regions of the polymer, improving ionic conductivity (1.4 × 10−4 S cm^{-1} at 30 °C). The incorporation of nanosized SiO_2 into PIL-based GPEs enhances mechanical properties while maintaining high ionic conductivity (F. Zhang *et al.*, 2020).

However, achieving both high ionic conductivity and robust mechanical strength in PIL-based electrolytes remains a challenge. High IL content, necessary for sufficient conductivity, often compromises mechanical integrity. Researchers address this by exploring cross-linking PILs, creating block/grafted copolymers, and incorporating inorganic additives (Niu *et al.*, 2021). Another promising avenue is the development of self-healing PIL-based electrolytes (R. Li *et al.*, 2022; Zhu *et al.*, 2022). These materials, like the imidazole-based PIL ion-conductive elastomer or PIL hydrogels incorporating choline, amino acids, and acrylamide, exhibit remarkable self-repair capabilities, ensuring longevity and resilience within LIBs (Lebedeva, Kultin and Kustov, 2023).

PILs also find applications beyond electrolytes. Their potential as anode materials, exploiting their redox groups for high-capacity "superlithiation" effects, is being explored (Yeji Wang *et al.*, 2023). Additionally, fluorinated PILs are emerging as superior cathode binders in both lithium ion and lithium-air batteries, significantly improving device capacity, performance, and cyclic stability (Eftekhari, Liu and Chen, 2016; Vizintin *et al.*, 2018).

The continued development of PIL-based electrolytes with tailored properties holds immense promise for the advancement of LIBs. By addressing the conductivity-mechanical strength trade-off and exploring innovative applications, PILs are poised to play a pivotal role in enabling safer, more powerful, and versatile lithium ion batteries.

2.3 Choline-Based ILs in Electrochemical Applications

2.3.1 Introduction

The urgent pursuit of sustainable energy storage demands electrolytes that combine safety, high performance, and minimal environmental impact. Choline-based ionic liquids (ILs) offer a compelling avenue due to their biodegradability, structural tunability, cost effectiveness, and potential for tailored physicochemical properties. While these ILs have found success in diverse electrochemical applications, their exploration within lithium batteries remains relatively limited. This section aims to identify promising candidates from other fields, analyzing their characteristics, and evaluate their potential for adaptation as advanced lithium battery electrolytes. A summary table (Table 2)provides a consolidated overview of how choline-based ILs have been utilized in lithium batteries, supercapacitors, dye-sensitized solar cells, actuators, and organic transistor highlighting their performance and key insights.

2.3.2 Lithium ion batteries

Yong et al. in 2016 investigated the potential of choline-based ionic liquids (ILs) as enhanced safety electrolytes within high-voltage lithium ion batteries. The researchers synthesized three novel ILs, incorporating trimethylsilyl, allyl, and cynoethyl groups, focusing on thermal stability, conductivity, electrochemical windows, and compatibility with lithium salts. Notably, the trimethylsilylated choline-based IL (SN1IL-TFSI) demonstrated particularly promising results. When used in a $LiCoO_2$/graphite full cell, this electrolyte enabled excellent cycling performance (152 mAh g^{-1} capacity, 99% retention over 90 cycles at 4.4V) and showed significantly reduced flammability compared to commercial electrolytes. The authors attribute the improved performance to the formation of a passivation film on the LCO cathode, suggesting that choline-based ILs with tailored functional groups offer a compelling avenue for developing safer and more stable high-voltage lithium ion battery electrolytes (Yong *et al.*, 2016). Rocha et al. in 2012 investigated the development of choline carboxylate zwitterionic ILs through the esterification of choline salts with a range of anhydrides for electrochemical applications. The researchers successfully employed two halogen-free synthetic routes to produce zwitterionic ILs. These ILs demonstrated several desirable electrolyte properties: a stable liquid state at room temperature, robust thermal stability up to 224°C, and excellent ionic conductivities (8.66×10^{-5} S cm^{-1} at 25°C to 1.80×10^{-3} S cm^{-1} at 90°C). These findings highlight the potential of choline carboxylate zwitterionic ILs as safer, more sustainable, and high-performance electrolyte materials (Rocha *et al.*, 2012).

Studies done by Millia et al. and Moradi and Farzi collectively investigated the potential of choline chloride (ChCl)/ethylene glycol (EG) (EG/ChCl) deep eutectic solvents (DESs) as electrolyte materials for lithium ion batteries. Millia et al. demonstrate the feasibility of incorporating lithium salts (LiTFSI and $LiPF_6$) into EG/ChCl DESs, achieving promising ionic conductivity (7.95 mS cm^{-1} at room temperature) with the 0.5 M $LiPF_6$/EG:ChCl formulation. The employed DESs (EG/ChCl 3:1 mol/mol and lactic acid (LA)/ChCl 2:1 mol/mol) were prepared by gentle heating under stirring at 70 °C for 15 min. The corresponding individual components were in the appropriate molar ratio until a clear solution was obtained. This formulation with easy synthesis method highlights the potential of these DESs as inherently safer and more cost-effective electrolyte options compared to traditional flammable organic electrolytes (Millia *et al.*, 2018; Moradi and Farzi, 2022).

Moradi and Farzi delve deeper, examining the impact of varying $LiPF_6$ salt concentrations within the EG/ChCl DES. Findings reveals that increasing salt concentration leads to higher viscosity but decreased ionic conductivity. This emphasizes the critical need to optimize salt concentration for balanced electrolyte performance. Computational modelling (Molecular Dynamics, MD simulation) in this study further elucidates the microstructure of the electrolyte, offering insights into ion interactions and diffusion behaviour (Moradi and Farzi, 2022).

Zafarani-Moattar et al. (2021) explore the influence of two deep eutectic solvents (DESs), based on choline chloride/lactic acid (ChCl/LA) and choline chloride/malonic acid (ChCl/MA), on the properties of lithium perchlorate ($LiClO_4$)(common electrolytic salt in lithium batteries) solutions in propylene carbonate (PC) (commonly used electrolyte for lithium batteries). The researchers combined experimental measurements (density, viscosity, speed of sound) with computational analysis (Density Functional Theory, DFT) and electrochemical measurements (Linear sweep voltammetry, LSV) to understand how varying DES composition and temperature affect thermodynamic, transport, and electrochemical properties. Results indicate stronger interactions between $LiClO_4$ and the ChCl/LA DES compared to the ChCl/MA counterpart. The ChCl/LA system shows particularly encouraging results with lower viscosity, a wider electrochemical window, and stronger $LiClO_4$ interactions, making it a leading candidate for further exploration within battery electrolytes. The ChCl/LA (1.54V) system also demonstrates a wider electrochemical potential window than ChCl/ MA system (1.34V) (Zafarani-Moattar *et al.*, 2021).

DiPietro et al. in 2022 investigated the potential of deep eutectic solvents (DESs) as safer alternatives to traditional electrolytes in lithium ion batteries. Specifically, the study focuses on the role of lithium cations (Li^+) in a choline chloride/urea (ChCl:U) DES containing lithium chloride (LiCl) salt. Researchers employ a combination of experimental NMR techniques and polarizable molecular dynamics simulations to gain a comprehensive understanding of the structural and dynamic properties of this system. Key findings reveal that Li^+ cations strongly coordinate with chloride anions, forming $LiCl_3^{2-}$ units. These interactions disrupt the original hydrogen-bonding network within the DES and influence interactions between Li^+ and other components, namely urea. This ultimately affects the overall molecular organization and ion mobility within the system (Di Pietro *et al.*, 2022).

Importantly, the study also considers the influence of residual water, a common impurity in DESs due to absorption from air. Findings indicate that water can impact interactions within the electrolyte and its overall physicochemical properties. This research underscores the importance of understanding how Li^+ cations interact with DES components, as this knowledge is crucial for the design and optimization of DES based electrolytes. The observed changes in molecular structure, interactions, and dynamics directly influence critical electrolyte properties, including conductivity, viscosity, and electrochemical stability, which ultimately determine battery performance. Additionally, the highlighted sensitivity to water content suggests the need for careful control during the development and application of DES electrolytes (Di Pietro *et al.*, 2022).

2.3.2 Supercapacitor

Supercapacitors are energy storage devices renowned for their lightning-fast charging and discharging capabilities, incredible power output, and exceptionally long lifespans (Ko *et al.*, 2019). They achieve this by storing energy electrostatically through the formation of an electrical double layer – a thin separation of charge at the interface between a high-surface-area electrode and an electrolyte (Du *et al.*, 2022).

The electrolyte plays a central role in supercapacitor performance. It must have high ionic conductivity to enable the rapid movement of ions during charging and discharging. A wide electrochemical stability window is equally important, allowing the supercapacitor to operate at higher voltages and thus store more energy (Tu, Delmerico and McDaniel, 2020). Additionally, the electrolyte needs to be chemically compatible with the electrode materials to prevent degradation, ensuring its longevity (Joo, Han and Cho, 2020). Safety considerations demand a non-flammable, chemically stable electrolyte, especially for high-power applications. Finally, a wide operating temperature range is desirable for the electrolyte to function reliably in diverse environments (Asl *et al.*, 2022).

W. Wang et al. (2023) introduced a novel and promising approach to utilizing choline chloride-based deep eutectic solvents (DESs) as environmentally friendly and cost-effective electrolytes within activated carbon-based electrochemical capacitors (ECs). The researchers prepared two DESs: choline chloride/glycerol (ChCl/G) and choline chloride/propionic acid (ChCl/PA), investigating their viscosity, conductivity, and electrochemical performance across a range of temperatures (room temperature to 373 K). Key findings demonstrate that rising temperatures favorably decrease DES viscosity and increase conductivity. Importantly, the symmetric ECs assembled with these choline chloride-based electrolytes exhibit excellent stability at high operating voltages (up to 2.5 V). The ChCl/PA electrolyte, particularly at 373 K, delivers a high specific capacitance of 198 F g-1 (energy density of 110 Wh kg-1). Notably, the supercapacitor assembled with ChCl/PA demonstrates excellent cycle stability, retaining 93% of its capacity over 5000 cycles at 373 K (Weirong Wang, Sabugaa, *et al.*, 2023).

In another study Nikpour, Moosavi and Torkzadeh (2023) explored the potential of using biodegradable choline-based ionic liquids (CBILs) as sustainable electrolytes for MXene-based supercapacitors. The researchers specifically investigate two CBILs: choline salicylate ([Ch][Sa]) and choline beta-alanine ([Ch][β-Ala]). These CBILs are chosen for their eco-friendly nature and potential compatibility with MXene materials. The study employs molecular dynamics simulations to examine how these CBILs behave within MXene nanopores under varying pore sizes and surface charge densities. Key findings reveal that confinement within MXene pores significantly alters CBIL structure and dynamics, with molecular structure and surface charge playing crucial roles in electrolyte performance (Nikpour, Moosavi and Torkzadeh, 2023).

Zhong et al. (2020) investigated the development of novel deep eutectic solvent (DES) electrolytes for supercapacitors with a wide operating temperature range (−40 °C, −20 °C, RT, 40 °C, 60 °C, 80 °C, 100 °C, 115 °C) . The researchers focus on a DES formed by mixing choline chloride (ChCl) and ethylene glycol (EG) in varying ratios. Density functional theory (DFT) calculations aid in determining the most stable DES formulation. Key findings show that the ChCl:EG ratio significantly influences electrolyte viscosity, conductivity, and supercapacitor performance. The optimal 1:2 ratio DES exhibits superior performance, especially at elevated temperatures (115 °C), demonstrating its potential for wide-temperature-range supercapacitor applications (Zhong *et al.*, 2020).

Xu et al. (2021) investigated the precise design of deep eutectic solvents (DESs) for superior supercapacitor electrolyte performance. The researchers utilize choline chloride (ChCl) as the hydrogen bond acceptor (HBA) and explore variations in butanediol isomers (with different hydroxyl group positions) as the hydrogen bond donor (HBD). Key findings demonstrate that the hydroxyl group positions in butanediols significantly influence the DES structure, properties, and ultimate supercapacitor performance. Among the studied systems, the

DES formed with ChCl and 1,2-butanediol (where the hydroxyl groups are closest) exhibits the highest conductivity (1.26 mS cm^{-1}) and lowest viscosity (56.99 cP). Importantly, density functional theory (DFT) calculations elucidate the hydrogen bonding mechanisms within the DESs, validating experimental observations. The ChCl/1,2-butanediol DES, when used as an electrolyte in an activated carbon (AC) based supercapacitor, demonstrates a voltage window of 2 V, a high specific capacitance of 116 Fg^{-1}, and an energy density of 16.14 Wh kg^{-1}. This work highlights the potential of tailored DES systems and emphasizes the value of combining DFT modeling with experimental studies for rational electrolyte design (Xu *et al.*, 2021).

Wang and Béguin (2022) investigated the development of a low-temperature electric double-layer capacitor (EDLC) utilizing an eco-friendly and cost-effective electrolyte based on aqueous choline bis(trifluoromethylsulfonyl)imide (ChTFSI). To enable low-temperature operation, the researchers employ a cosolvent strategy, optimizing the mixture of water with methanol. This ChTFSI/methanol-water electrolyte demonstrates excellent conductivity (22.8 mS cm^{-1} at 20°C and 3.1 mS cm^{-1} at -30°C). Importantly, the EDLC demonstrates stable operation down to -30 °C with a cell voltage of 1.6 V (Wang and Béguin, 2022).

Recent study done by Mirzaei-Saatlo et al. in 2024 introduces a novel phase change ionogel electrolyte designed for use in flexible supercapacitors. This electrolyte is synthesized from 2-hydroxyethyl cellulose (HEC) and choline formate ([Ch]Fo) ionic liquid. Key findings reveal that this [Ch]Fo/HEC ionogel exhibits a high latent heat, thermal stability exceeding 95%, and impressive mechanical strength (tensile strength of 1.69 MPa). These properties, combined with strong water retention, facilitate the development of high-performance supercapacitors. When integrated into a flexible supercapacitor with a poly(aniline-co-4-nitroaniline) electrode, the system achieves a high specific capacitance of 594 Fg^{-1} at 0.5 A g^{-1}. Additionally, the device demonstrates a maximum energy density of 29.7 Wh kg-1 (at 0.5 A g^{-1}) and a power density of 1500 W kg^{-1} (at 5 A g^{-1}) (Mirzaei-Saatlo *et al.*, 2024).

2.3.3 Dye-sensitized solar cells (DSSCs)

DSSCs offer a unique alternative to conventional silicon solar cells. They harness a photoelectrochemical process, somewhat similar to photosynthesis, to convert sunlight into electricity (Sie and Ngaini, 2017). A key component is a special dye-covered electrode (often made of titanium dioxide), a counter electrode, and an electrolyte solution that fills the space between them (D and Pesala, 2019). When sunlight hits the dye, electrons get excited and jump into the titanium dioxide, starting an electrical current (Rahman *et al.*, 2021).

The electrolyte plays a vital role in keeping the DSSC running. It transports charge, ensuring that the dye molecules are continuously regenerated to absorb more light. For this to work, the electrolyte needs high ionic conductivity for fast electron transfer (Rani *et al.*, 2021). Additionally, it must be chemically compatible with the dye and electrodes to prevent degradation (Han *et al.*, 2015). The specific chemicals in the electrolyte are chosen for their ability to efficiently regenerate the dye molecules. Finally, a good electrolyte for DSSCs should also be non-volatile (to prevent leakage) and function across a wide range of temperatures (Muchuweni, Martincigh and Nyamori, 2020).

Ku and Lu introduced in 2011 a novel room temperature ionic liquid (RTIL) based on acetylcholine iodide and ethylene glycol (ACI/EG) for use in low volatility DSSC electrolytes. This RTIL offers advantages in terms of cost-effectiveness, ease of preparation, and environmental friendliness compared to conventional imidazolium-based RTILs. The ACI/EG electrolyte demonstrates low volatility and suitable properties like viscosity and conductivity. Importantly, a DSSC using this electrolyte achieves a power conversion efficiency of 3.36%, which is 82% of the efficiency obtained with a standard acetonitrile-based electrolyte (Ku and Lu, 2011).

Boldrini et al. in 2017 pioneered the use of a choline chloride-based DES (ChCl/glycerol, 1:2 mol/mol) as an environmentally friendly electrolyte for DSSCs. The researchers meticulously optimize the DES composition by incorporating water (40% water content) to achieve compatibility with a hydrophilic organic sensitizer. This strategic approach results in a DSSC demonstrating power conversion efficiency (PCE) on par with DSSCs utilizing traditional, yet often hazardous, organic solvents. Importantly, this work highlights the potential of choline based DESs to replace toxic and volatile electrolytes, paving the way for more sustainable solar cell technologies (Boldrini *et al.*, 2017).

In another study Nyuyen et al. (2019) investigated the integration of a choline chloride-phenol DES into DSSC electrolytes to enhance sustainability and long-term performance. The researchers strategically blend the DES with acetonitrile, a conventional electrolyte solvent. Importantly, the DES-based electrolyte system demonstrates improved stability, with the efficiency of a DSSC containing 20% DES increasing from 6.92% to 7.75% after 1300 hours of operation. Density functional theory (DFT) calculations reveal that the choline and phenyl groups within the DES co-adsorb onto the TiO_2 surface, contributing to the observed stabilization (Nguyen *et al.*, 2019).

By the same research group, Nguyen et al. (2021) investigated the development of eco-friendly and cost-effective DES electrolytes for use in DSSCs. Two novel DESs are formulated: one composed of choline chloride and urea (DES-CU) and another with choline chloride and ethylene glycol (DES-CE). Crucially, these DESs offer advantages including ease of large-scale production, non-toxicity, and biodegradability. Importantly, the DES-based DSCs demonstrate conversion efficiencies comparable to those using a standard ionic liquid electrolyte. Electrochemical impedance spectroscopy reveals that the DES molecular structures differentially influence performance parameters: DES-CU enhances open-circuit voltage (VOC), while DES-CE increases short-circuit current (ISC) (Nguyen *et al.*, 2021).

Boogaart, Essner, and Baker (2022) investigated the impact of halide selection on the performance of choline halide/guanidinium thiocyanate deep eutectic solvent (DES) electrolytes in dye-sensitized solar cells (DSCs). Three DESs are examined: guaniline-Cl, guaniline-Br, and guaniline-I. Key findings reveal a general trend of increasing device performance with decreasing hydrogen bond affinity of the halide (Cl- < Br- < I-). Importantly, the guaniline-I system demonstrates significantly higher photocurrents compared to the other DESs. Interestingly, the guaniline-I DES, due to its incorporated iodide, can function effectively as the sole iodide source for the redox couple. This simplifies the electrolyte composition and allows for comparable performance to systems with supplemental inorganic iodide, despite differences in viscosity (Boogaart, Essner and Baker, 2022).

Heydari Dokoohaki, Mohammadpour and Zolghadr (2011) investigated the performance of a choline chloride-ethylene glycol (ChCl-EG) DES electrolyte in DSSC. The researchers explore the influence of different iodide salt additives (potassium iodide; KI, and an IL, 1-etyl-3-methylimidazolium iodide; EmimI) on electrolyte properties and DSSC performance. Importantly, the presence of KI significantly enhances DSSC efficiency, despite increasing electrolyte viscosity. Electrochemical analysis reveals that KI-containing DES systems demonstrate higher electron lifetimes and charge recombination resistance. Complementary molecular dynamics simulations illustrate that K^+ cations strongly interact with the TiO_2 surface, potentially contributing to the observed performance gains (Heydari Dokoohaki, Mohammadpour and Zolghadr, 2021).

2.3.4 Actuator

An actuator is a device that turns energy into controlled motion. Actuators come in many forms, including hydraulic, pneumatic, electric motors, and more(Xiang *et al.*, 2017). Electrolytes are not essential for all types of actuators, but they play a critical role within electrochemical actuators. These actuators use electrochemical reactions to make materials expand or contract, which causes movement (Ling *et al.*, 2022). The electrolyte provides the ions needed for these reactions and acts as the medium for ion transport within the actuator. Specific examples of electrolyte-based actuators include conducting polymer actuators and ionic polymer-metal composites (IPMCs)(Luqman *et al.*, 2022).

Elhi et al. (2020) developed biofriendly electroactive polymer actuators using choline-based ionic liquids (ILs) as electrolytes. Six choline ILs are selected for their low toxicity and promising suitability for biomedical applications. Importantly, the study evaluates the toxicity of selected choline ILs against various microbial cultures and human cells, demonstrating their minimal harmful effects. The fabricated trilayer polypyrrole-PVdF actuators using these choline ILs demonstrate excellent performance, with choline acetate even outperforming an imidazolium-based reference system (Elhi, Priks, *et al.*, 2020).

By the same research group Elhi et al investigated fully biocompatible soft actuators composed of polypyrrole-gelatin trilayers using choline-based ionic liquids (ILs) as electrolytes. Choline acetate and choline isobutyrate actuators demonstrate superior performance, including the highest strain difference and strain difference to charge density ratio, even outperforming actuators using an imidazolium-based IL and PVdF membrane. Notably, the ionic conductivity (Table 1) in all of the six choline based ILs in gelatin show great potential in lithium ion batteries. Also, these actuators exhibit no adverse effects in bacterial testing, confirming their biofriendly nature. To understand this behavior, molecular dynamics simulations and density functional theory calculations reveal significant cation-cation clustering within the ILs. The height of the cation-cation correlation peak within the simulations strongly correlates with the experimentally observed actuator strain difference (Elhi, Karu, *et al.*, 2020).

Table 1. Ionic conductivities of Choline based IL in gelatin and its ionic conductivities under atmospheric conditions (Elhi, Priks, *et al.*, 2020).

Ionic Liquid	Ionic conductivities $\kappa/\mathrm{mS\cdot cm^{-1}}$
Choline acetate ([Ch][Ac])	0.48
Choline isobutyrate ([Ch][Ib])	0.06
Choline isovalerate ([Ch][Iv])	0.06
Choline 2-methylbutyrate ([Ch][2 mb])	0.01
Choline malonate ([Ch][Mal])	0.05
Choline glutarate ([Ch][Glu])	0.00

2.3.5 Organic Transistor

Organic transistors are a type of transistor that utilizes organic semiconductors (like special polymers or small molecules) instead of the traditional silicon (Uno *et al.*, 2015). This opens up possibilities for flexible, potentially lower-cost, and even transparent electronics. They can be made on bendable substrates, leading to exciting possibilities like roll-up displays (Ganesan, Tsao and Gao, 2021). However, organic transistors currently have limitations compared to silicon transistors, including generally lower performance (speed) and potential issues with long-term stability (Y. Xu *et al.*, 2015). Nonetheless, they hold promise for specific applications where flexibility and low cost are prioritized. Electrolytes can play a crucial role in specific types of organic transistors, such as electrolyte-gated transistors, where they modulate the conductivity of the organic semiconductor, enabling transistor operation (Giovannitti *et al.*, 2016).

Jo et al in 2020 studied the development of biocompatible and biodegradable organic transistors enabled by a novel solid-state electrolyte. The electrolyte ingeniously combines levan polysaccharide, a biocompatible polysaccharide, with a custom-synthesized choline-based ionic liquid. This ionic liquid, formed by reacting choline with malic acid, is crucial for achieving the electrolyte's desirable mechanical and electrochemical properties. The electrolyte exhibits a large specific capacitance (≈40 μF cm^{-2} at 10 Hz), flexibility, and transparency, contributing to the development of low-voltage, mechanically robust organic transistors. Importantly, the study demonstrates the potential of these transistors for direct integration with biological systems. The researchers successfully use the devices for high-quality electrocardiogram (ECG) recordings on both human skin and a rat's heart. The solid-state nature of the electrolyte facilitates conformal contact with biological surfaces, enhancing signal acquisition. Additionally, the biodegradability of the electrolyte's components highlights its potential for transient biomedical monitoring applications (Jo *et al.*, 2020).

Table 2. Summary of various choline ILs used in different electrochemical applications.

Application	Choline ILs Used	Key Insights	References
Lithium batteries	- (2-hydroxyethyl)trimethylammonium bis(trifluoromethanesulfonyl)im-ide (N1IL-TFSI) - (2-trimethylsililoxyethyl)trimethylammonium bis(trifluoromethanesulf-onyl)imide (SN1IL-TFSI) - (2-allyloxyethyl)trimethylammonium bis(trifluoromethanesulfonyl)imi-de (AN1IL-TFSI)	**Choline-based ILs as safer electrolytes:** This study demonstrates the feasibility of designing choline-based ionic liquids that are less flammable than traditional electrolytes, improving the safety of high-voltage lithium ion batteries. **Functional groups matter:** The specific functional groups attached to the choline-based IL significantly impact electrolyte properties. Trimethylsilyl groups appear to enhance cell performance and safety. **Passivation film formation:** The improved cycling stability in the $LiCoO_2$/graphite cell suggests that the SN1IL-TFSI electrolyte assists in forming a protective passivation layer on the cathode, extending battery life. **High-voltage compatibility:** The research indicates that these designed ionic liquids have the potential to work well in high-voltage battery systems, which could increase energy density.	(Yong *et al.*, 2016)
	- succinylmonocholine lithium bis(trifluoromethylsulfonyl)imide - glutarylmonocholine lithium bis(trifluoromethylsulfonyl)imide - 3-methyglutarylmonocholine lithium bis(trifluoromethylsulfonyl)imide	**Halogen-free synthesis:** The development of halogen-free synthetic methods for ILs offers a more environmentally conscious approach to electrolyte production. **Room-temperature liquid state:** Zwitterionic ILs that remain liquid at room temperature offer practical advantages for handling and integration into electrochemical devices. **High thermal stability:** Good thermal stability is essential for electrolyte safety and longevity in battery applications. **Promising conductivity:** The demonstrated ionic conductivity of these ILs suggests their suitability for applications where efficient ion transport is critical for performance.	(Rocha *et al.*, 2012)
	choline chloride (ChCl)/ethylene glycol (EG) (EG/ChCl) deep eutectic solvents (DESs)	**Choline chloride/EG DES as a promising electrolyte base:** These studies collectively support the viability of choline chloride/ethylene glycol DESs as a foundation for lithium ion battery electrolytes.	(Moradi and Farzi, 2022)

		Safety and cost advantages: EG/ChCl DESs offer the potential for improved safety due to their non-volatility and non-flammability, along with potential cost benefits. **Salt concentration is key:** Optimizing the concentration of lithium salts within the DES is crucial for achieving a desirable balance between ionic conductivity and viscosity. **Value of computational modeling:** Molecular simulations, as demonstrated in Research 2, provide valuable insights into the microstructure and ion dynamics within DES electrolytes, aiding in the rational design and improvement of their properties.	(Millia *et al.*, 2018)
	choline chloride/urea (ChCl:U)	**Presences of Li^+**: The presence of lithium cations in the DES significantly alters the intermolecular interactions and dynamics, influencing properties important for electrolyte performance. **Lithium coordination:** Lithium cations strongly coordinate with chloride anions, forming LiCl3(2-) units. This affects the hydrogen-bond network and molecular organization within the DES. **Interaction:** Lithium cations interact with other components, particularly urea, influencing ion mobility and solvation behavior. **Effect of water:** Residual water, commonly present in DESs due to absorption from air, can impact the interactions within the electrolyte and its physicochemical properties.	(Di Pietro *et al.*, 2022)
	choline chloride/lactic acid (ChCl/LA) and choline chloride/malonic acid (ChCl/MA)	**DES composition matters:** The specific choice of DES (ChCl/LA vs. ChCl/MA) significantly influences interactions with the LiClO4 electrolyte, affecting performance properties. **Potential for electrolyte development:** Both DES systems show promise for lithium ion battery electrolytes due to low viscosity, favorable interactions with LiClO4, and suitable electrochemical stability windows. **ChCl/LA advantage:** The ChCl/LA system shows particularly encouraging results with lower viscosity, a wider electrochemical window, and stronger LiClO4 interactions, making it a leading candidate for further exploration within battery electrolytes. **Combined experimental/theoretical approach:** This study highlights the value of combining experimental characterization with computational modeling (DFT) to gain deeper insights into DES-based electrolyte behavior.	(Zafarani-Moattar *et al.*, 2021)

Supercapacitor	choline chloride/glycerol (ChCl/G) and choline chloride/propionic acid (ChCl/PA)	**Ultra-fast Charging/Discharging:** Energy storage and release happen in seconds, not hours! **High Power Density:** They can deliver bursts of power far exceeding batteries. **Long Cycle Life:** They can endure hundreds of thousands of charge-discharge cycles.	(Weirong Wang, Sabugaa, *et al.*, 2023)
	choline salicylate ([Ch][Sa]) and choline beta-alanine ([Ch][β-Ala])	**Focus on biodegradable choline electrolytes:** The study highlights the potential of CBILs as greener alternatives for energy storage applications. **Importance of molecular structure:** The specific choice of CBIL ([Ch][Sa] vs. [Ch][β-Ala]) impacts interactions with MXene and overall electrolyte behavior. **Confinement effects:** MXene pore size significantly influences CBIL structure and dynamics. **Electrostatic control:** Surface charge density on MXene electrodes offers a potential mechanism to fine-tune electrolyte properties.	(Nikpour, Moosavi and Torkzadeh, 2023)
	choline chloride (ChCl) and ethylene glycol (EG)	**ChCl/EG DES for wide temperature range:** This DES offers a promising electrolyte solution capable of operating effectively across a wide temperature range (-40 °C to 115 °C). **Importance of composition:** The molar ratio of ChCl to EG is crucial for optimizing electrolyte viscosity, conductivity, and overall supercapacitor performance. **DFT for optimization:** Density functional theory calculations help identify the most stable and promising DES formulation. **High-performance at elevated temperatures:** The DES-1-2 (1:2 ratio) demonstrates particularly strong performance at high temperatures, offering significant energy density.	(Zhong *et al.*, 2020)
	ChCl with 1, 2-butanediol, 1, 3-butanediol and 1, 4-butanediol	**Hydroxyl group position is crucial:** The placement of hydroxyl groups on the butanediol molecule significantly impacts DES properties and supercapacitor performance. **Optimal DES for supercapacitors:** The ChCl/1,2-butanediol combination yields a DES with the highest conductivity, lowest viscosity, and superior electrochemical performance. **DFT validation:** Density functional theory calculations provide insights into hydrogen bonding mechanisms, supporting experimental findings.	(Xu *et al.*, 2021)

	choline bis(trifluoromethylsulfonyl)imide (ChTFSI)	**Choline-based electrolyte for low temperatures:** Aqueous ChTFSI offers advantages in safety, cost, and environmental impact for EDLC electrolytes. **Cosolvent enables performance:** The optimized methanol-water mixture lowers the electrolyte's freezing point and enhances conductivity, facilitating low-temperature operation. **Impressive performance:** The EDLC achieves stable operation down to -30°C with a cell voltage of 1.6V. **Promising for eco-friendly energy storage:** This work demonstrates the potential of choline-based aqueous electrolytes with cosolvents for the development of safer, low-temperature EDLCs.	(Wang and Béguin, 2022)
	2-hydroxyethyl cellulose (HEC) and choline formate ([Ch]Fo)	**Choline formate-based ionogel:** Choline formate combined with HEC forms a unique phase change ionogel electrolyte for flexible supercapacitors. **Thermal and mechanical advantages:** The electrolyte offers high latent heat, strong thermal stability, and excellent mechanical strength, making it well-suited for flexible devices. **Impressive electrochemical performance:** The flexible supercapacitor demonstrates high specific capacitance, energy density, and power density. **Potential for green energy storage:** This research highlights the potential of utilizing choline-based phase change ionogels within flexible energy storage devices.	(Mirzaei-Saatlo *et al.*, 2024)
Dye-Sensitized Solar Cells	acetylcholine iodide and ethylene glycol (ACI/EG)	**Choline-based RTIL for safer, low volatility electrolytes:** This study demonstrates the potential of using acetylcholine iodide as a component in RTIL electrolytes for DSSCs. **Advantages over traditional RTILs:** The ACI/EG system offers benefits in cost, preparation, and environmental impact. **Promising performance:** The DSSC employing the ACI/EG electrolyte demonstrates good power conversion efficiency. **Potential for further development:** This work encourages further research into the development and optimization of choline-based RTILs for DSSC applications.	(Ku and Lu, 2011)

	ChCl/glycerol, 1:2 mol/mol	**Choline chloride-based DES electrolyte:** This DES presents a greener and safer alternative for DSSCs. **Importance of water content:** The optimized electrolyte incorporates 40% water, crucial for compatibility and performance. **Synergy with sensitizer:** By employing a hydrophilic organic sensitizer, the researchers effectively exploit the aqueous DES electrolyte. **Significant performance:** The DES-based DSSC achieves PCE comparable to DSSCs using conventional, often toxic, electrolytes.	(Boldrini *et al.*, 2017)
	choline chloride-phenol	**Choline-based DES for enhanced DSC stability:** The addition of a choline chloride-phenol DES to a conventional electrolyte significantly improves long-term DSC performance. **Optimal DES integration:** A 20% DES mixture with acetonitrile demonstrates the best balance of initial performance and long-term stability. **DFT insights:** Density functional theory calculations suggest that the co-adsorption of choline and phenyl groups from the DES onto TiO2 contributes to enhanced stability. **Promising for eco-friendly DSCs:** This study highlights the potential of DESs as greener and performance-enhancing electrolyte components within DSCs.	(Nguyen *et al.*, 2019)
	choline chloride and urea (DES-CU) and choline chloride and ethylene glycol (DES-CE)	**Choline-based DESs for sustainable DSCs:** The study introduces DESs containing choline chloride as safer, more environmentally friendly, and cost-effective electrolyte options for DSCs. **Comparable performance:** The DES-based DSCs achieve conversion efficiencies similar to a benchmark ionic liquid electrolyte. **Tailored functionality:** The choice of DES (DES-CU vs. DES-CE) allows for tuning specific performance parameters like VOC or ISC. **Potential for large-scale production:** The simple synthesis and desirable properties of these DESs suggest their suitability for scalable DSC manufacturing.	(Nguyen *et al.*, 2021)
	guaniline-Cl, guaniline-Br, and guaniline-I	**Halide choice matters:** In choline-based DES electrolytes, weaker hydrogen bond affinity of the halide (e.g., iodide) correlates with lower viscosity and enhanced DSC performance. **Guanidine-I advantages:** This DES demonstrates significantly higher photocurrents and offers a promising avenue for further electrolyte development.	(Boogaart, Essner and Baker, 2022)

		Self-contained electrolyte: The iodide within guanidine-I can act as the sole iodide source, simplifying electrolyte design and offering performance advantages. **Future directions:** The research suggests focusing on iodide-containing DESs and tailoring DES components for lower viscosity and integration of electrolyte additives.	
	choline chloride-ethylene glycol (ChCl-EG)	**Choline-based DES as a DSSC electrolyte:** ChCl-EG DES offers a potential electrolyte solvent for DSSCs. **KI provides performance advantages:** The addition of KI improves DSSC efficiency, likely due to enhanced electron lifetime, charge recombination resistance, and favorable interactions with TiO2. **Understanding the trade-offs:** While KI increases electrolyte viscosity, the performance benefits appear to outweigh the drawbacks. **Insights from molecular simulations:** Simulations reveal strong K+ interactions with the TiO2 surface and provide insights into the solvation of ions within the DES electrolyte.	(Heydari Dokoohaki, Mohammadpour and Zolghadr, 2021)
Actuator	Choline acetate ([Ch][Ac]) Choline isobutyrate ([Ch][Ib]) Choline isovalerate ([Ch][Iv]) Choline 2-methylbutyrate ([Ch][2 mb]) Choline malonate ([Ch][Mal]) Choline glutarate ([Ch][Glu]) Choline citrate ([Ch][Cit])	**Biofriendly actuators:** This study demonstrates the feasibility of creating electroactive polymer actuators with enhanced biocompatibility using carefully selected components. **Choline ILs for safer devices:** The use of choline ILs with demonstrated low toxicity significantly improves the safety profile of the actuators. **Toxicity evaluation:** Comprehensive testing against various microbial cultures and HeLa cells confirms the low toxicity of the selected choline ILs. **Excellent performance:** The choline IL-based actuators show promising actuation properties, with choline acetate surpassing the benchmark imidazolium-based IL.	(Elhi, Priks, *et al.*, 2020)
		Fully non-toxic soft actuators: This study demonstrates the feasibility of creating high-performance, biocompatible soft actuators using gelatin and choline ILs. **Choline ILs enhance performance:** Actuators incorporating choline acetate and choline isobutyrate outperform those using traditional imidazolium-based ILs and PVdF membranes. **Biocompatibility confirmed:** Testing against *E.coli* demonstrates the non-toxic nature of the polypyrrole-gelatin/choline IL actuators.	(Elhi, Karu, *et al.*, 2020)

		Computational insights: Simulations reveal that cation-cation clustering within the choline ILs strongly influences actuator performance.	
Organic Transistor	choline malic acid[Ch][MA]	**Bioderived electrolyte for bioelectronics:** The research presents a solid-state electrolyte composed of levan polysaccharide and a choline-based ionic liquid, designed for safe integration with biological systems. **Choline-based IL's role:** The choline-based ionic liquid enhances electrolyte flexibility and electrochemical performance, enabling low-voltage and robust organic transistors. **Direct bio-integration:** The electrolyte's properties allow for direct contact with biological tissues, facilitating high-quality ECG signal acquisition. **Biocompatibility and biodegradability:** The edible nature of the electrolyte components ensures biocompatibility and the potential for safe resorption within the body.	(Jo *et al.*, 2020)

2.3.4 Electrochemical Potential Windows (EPWs) of Choline Based DES.

The EPWs of an electrolyte defines the voltage range where it stays chemically stable. It acts like the voltage "safe zone" for the electrolyte. Exceeding this window can trigger electrolyte breakdown, potentially affecting performance, longevity, and even safety in energy storage devices (Mousavi *et al.*, 2015). The EPWs depends heavily on the specific electrolyte ingredients (salts, solvents) and can be influenced by factors like electrode interactions and temperature (Lian *et al.*, 2018). Researchers aim to design electrolytes with wide EPWs, as this allows for higher voltage operation in devices like batteries and supercapacitors, leading to increased energy storage capacity (Guan, Croft and Grant, 2022). Li et al. (2016) did a comprehensive study investigating the EPWs (Table 3) of various choline based deep eutectic solvents (DESs), aiming to characterize their electrochemical stability. The researchers employ cyclic voltammetry to examine choline based DESs, exploring the influence of structural factors on the EPWs. For DESs, the research demonstrates that the choice of hydrogen bond donor (HBD) significantly impacts the EPW in choline chloride (ChCl) based systems. The DES composed of ChCl and methyl urea exhibits the largest potential window (4.72 V). The cation limit of ChCl based DES varied depending on the HBD. The limit increased in the following order: oxalic acid < glycerol < ethanediol < malonic acid < butanediol < xylitol < urea < methyl urea. Interestingly, ChI-based DESs with oxalic acid displayed the widest potential window (2.76 V), narrower than the smallest window in ChCl-based systems. This points to a complex interplay between ChCl stability and HBD-HBA interactions in determining electrochemical windows. (Li *et al.*, 2016).

These findings offer valuable guidance for selecting choline based DESs with desirable electrochemical properties for various applications. The researcher's systematic approach highlights the interplay of structural components, electrode choice, and sensitivity to water content. This work provides a strong foundation for designing and optimising electrolytes in electrochemical devices requiring wide electrochemical potential windows for enhanced performance and stability (Li *et al.*, 2016).

Table 3.The cathodic (E_{CL}), anodic limits (E_{AL}) and EPWs of ChCl and methyl urea based DESs measured by utilizing CV technique on the glassy carbon working electrode at 100 mV/s. (Li *et al.*, 2016)

DESs	E_{AL}	E_{CL}	EPWs
ChCl+oxalic acid	-0.92	1.24	2.16
ChCl+glycerol	-2.21	1.38	3.59
ChCl+ethanediol	-2.35	1.26	3.61
ChCl+malonic acid	-2.55	1.70	4.25
ChCl+1,4-butanediol	-2.57	1.33	3.90
ChCl+xylitol	-2.67	1.66	4.33
ChCl+urea	-2.75	1.54	4.29
ChCl+methyl urea	-3.06	1.66	4.72
$ChNO_3$+methyl urea	-1.76	0.76	2.52
$ChClO_4$+methyl urea	-2.49	2.04	4.53
$ChBF_4$+methyl urea	-1.66	2.03	3.69
ChBr+glycerol	-2.36	1.16	3.52
ChBr+ethanediol	-1.35	0.77	2.12
ChBr+malonic acid	-2.38	1.03	2.41
ChBr+butanediol	-1.14	0.58	1.72
ChBr+xylitol	-2.67	1.66	4.33
ChBr+urea	-2.09	1.23	3.32

ChBr+methyl urea	-1.76	0.82	2.58
ChI+methyl urea	-1.73	0.36	2.09
ChI+oxalic acid	-2.32	0.44	2.76
ChI+ethanediol	-2.38	0.30	2.68
ChI+glycerol	-2.17	0.42	2.59
ChI+urea	-0.89	0.36	1.25

2.3.5 Potential Candidate for Choline Based Electrolyte.

Despite the limited exploration of choline-based ionic liquids (ILs) specifically within lithium batteries, their diverse applications in other fields offer a unique starting point for identifying promising electrolyte candidates. This section leverages a comprehensive table (Table 4) compiled by Gadilohar and Shankarling, which meticulously categorizes various choline based ILs and their existing functionalities in areas such as lubrication, organic synthesis, and biomass pretreatment. Importantly, the physicochemical properties that make these ILs successful in their respective fields could hold untapped potential for lithium battery electrolytes. For instance, ILs demonstrating high thermal stability or tailored solvent properties might translate well to battery applications. By systematically evaluating the ILs listed in the table and considering the specific requirements for lithium battery electrolytes, we can uncover hidden gems within this pool of choline-based materials (Gadilohar and Shankarling, 2017).

Table 4. List of choline based ILs and its remarks and application. (adapted from the work of Gadilohar and Shankarling) (Gadilohar and Shankarling, 2017).

No	Choline Task-specific ionic liquids (TSIL)	Remarks/applications
1	Choline salicylate (BASIONIC™ FS 65)	Drilling fluids

No	Choline Task-specific ionic liquids (TSIL)	Remarks/applications
		Additive in pharma. prod. Additive for polymerization of polyoxymethylene copolymers
2	Choline bis(salicyl)borate	High performance lubricant Additives for lubricant Extraction of metal ions of rare earth elements.
3	Choline acetate (BASIONIC™ FS 85)	Potential electrolytes Free-radical polymerization
4	Choline itaconate	Potential electrolytes Free-radical polymerization
5	Choline propionate	Basic TSIL
6	Choline butanoate	Basic TSIL
7	Choline pivalate	Basic TSIL
8	Choline hexanoate	Basic TSIL
9	Choline lactate	Basic TSIL
10	Choline persulfate	Selective and rapid oxidation of alcohols Sonochemical selective oxidation Synthesis of Betti bases and gem-bisamides Concentrated solar radiation aided oxidation of alcohol
11	Choline bisulfate	Acid catalyzed reaction 3,4-dihydro-2(H)-pyrimidinones Conversion of agar into mixed sugars
12	Choline lauryl sulphate	Base catalyzed reaction Choline soaps Green surfactant
13	Choline docusate	Green surfactant
14	Choline hydroxide	Base catalyzed reaction Synthesis of dimethyl carbonate Synthesis of 4-aminoquinoline-2-ones Ring opening reaction of cyclohexane oxide Synthesis of tetrahydrobenzo[*b*]pyran Knoevenagel–Michael reaction Synthesis of propylene glycol ether Aldol reaction
15	Choline 2-naphthoxyacetate	Basic TSIL
16	Choline anthracene-9-carboxylate	Basic TSIL
17	Choline triflate	Acidic TSIL
18	Choline acesulfamates	Hydrophobic IL with low toxicity
19	Choline saccharinate	Hydrophobic IL with low toxicity Azo dyes degradation

No	Choline Task-specific ionic liquids (TSIL)	Remarks/applications
20	Choline dimethyl phosphate	Free-radical polymerization
21	Choline bis(trifluoromethylsulfonyl)amide	Proton transport Extraction of neodymium(iii)
22	Choline monohydrophosphate	Free-radical polymerization
23	Choline dihydrophosphate	Real-world green biological buffers For antibody storage and transportation.
24	Choline cyclopentane carboxylate	Basic TSIL
25	Choline cyclopentyl acetate	Basic TSIL
26	Choline cyclohexane carboxylate	Basic TSIL
27	Choline cholanate	Basic TSIL
28	Choline deoxycholate	Basic TSIL
29	Choline lithocholate	Basic TSIL
30	Choline benzoate	Basic TSIL
31	Choline p-toluene sulfonate	Basic TSIL
32	Choline citrate	Basic TSIL
33	Choline fumarate	Basic TSIL
34	Choline glutarate	Basic TSIL
35	Choline tartarate	Basic TSIL
36	Choline decanoate	Basic TSIL
37	Choline glycolate	Basic TSIL
38	Choline 3-cyclohexyl propionate	Basic TSIL
39	Choline oleate	As ionic grease lubricants
40	Choline thioglycolate	For dissolution of feather keratin
41	Choline glycinate	Application in the pre-treatment of biomass CO_2 capture
42	Choline l-alaninate	Application in the pre-treatment of biomass
43	Choline l-serinate	Application in the pre-treatment of biomass
44	Choline l-prolinate	Application in the pre-treatment of biomass
45	Choline l-isoleucinate	Application in the pre-treatment of biomass
46	Choline l-lycinate	Application in the pretreatment of biomass
47	Choline l-cysteinate	Application in the pretreatment of biomass
48	Choline l-methioninate	Application in the pretreatment of biomass
49	Choline l-histidinate	Application in the pretreatment of biomass

No	Choline Task-specific ionic liquids (TSIL)	Remarks/applications
50	Choline l-phenyl alaninate	Application in the pretreatment of biomass
51	Choline l-leucinate	Application in the pretreatment of biomass
52	Choline l-aspartate	Application in the pretreatment of biomass
53	Choline l-phenyl alaninate	Application in the pretreatment of biomass
54	Choline l-tyrosinate	Application in the pretreatment of biomass
55	Choline l-valinate	Application in the pretreatment of biomass
56	Choline l-nor valinate	Application in the pretreatment of biomass
57	Choline l-nor leucinate	Application in the pretreatment of biomass
58	Choline tiglate	Application in the pretreatment of biomass
59	Choline H-maleate	Application in the pretreatment of biomass
60	Choline H-malate	Basic TSIL
61	Choline H-succinate	Application in the pretreatment of biomass
62	Choline octanoate	Basic TSIL
63	Choline formate	Polymerization of vinyl and methacrylic monomers
64	Choline argininate	Pretreatment solvents for grass lignocelluloses
65	Choline trifluoroacetate	Basic TSIL
66	Choline methanesulfonate	Acidic TSIL
67	Choline oxopentanoate	Basic TSIL
68	Choline oligoether carboxylate	Oligoether TSIL
69	Choline purpurin-18	Photosensitizers Anticancer agents
70	Choline chloride:2 urea	Halogenation reaction Synthesis of tricyanovinylated aromatics Perkin reaction β-Hydroxy functionalized derivatives Synthesis of 1,3,4-oxadiazole β-Hydroxy ketonic derivatives Synthesis of oxazoles Selective *N*–alkylation CSR assisted synthesis of DES
71	Choline chloride:malonic acid	Synthesis of novel 2,3-dihydroquinazolin-4(1H)-one derivatives
72	Choline chloride:oxalic acid	Acid-catalyzed transformation Direct C-3 alkenylation/alkylation of indoles
73	Choline chloride:citric acid	Acid-catalyzed transformation
74	Choline chloride in water	Coumarin synthesis by Knoevenagel condensation

No	Choline Task-specific ionic liquids (TSIL)	Remarks/applications
75	Choline chloride:urea:glycerol	Synthesis of *N*-aryl phthalimide
76	Choline chloride:zinc chloride	Kabachnik–Fields reaction Nucleophilic substitution reactions of alcohols Protection of carbonyls to 1,3-dioxolanes Synthesis of cyclic carbonates from CO_2
77	Choline chloride:tin chloride	Synthesis of phytosteryl esters One-pot synthesis of quinolone
78	Choline chloride:copper chloride	Synthesis of 1,2,3-triazoles
79	Choline chloride:chromium chloride	Esterification of formic and acetic acid
80	Choline chloride:urea:$AlCl_3$	Electrodeposition of aluminium on cathodes

2.3.6 Summary

This section of review has highlighted several classes of choline based ILs that hold potential for lithium battery electrolytes, particularly those exhibiting favorable conductivity and thermal stability. Key challenges include optimizing viscosity, electrolyte ratio and ensuring compatibility with lithium battery components. The findings emphasize the strong link between ILs structure and electrolyte performance. Additionally, investigating the formation of gel polymer electrolytes (GPEs) using the potential choline based ILs offers a promising avenue for further enhancing safety and electrochemical stability. The upcoming section will delve into the advantages of GPEs and explore their potential synergies with choline-based ILs for the development of next-generation lithium battery electrolyte.

2.4 Recent Advancement and Challenges in Polymer Electrolyte

2.4.1 Introduction

Solid polymer electrolytes (SPEs), formed by dissolving lithium salts within solid polymer matrices, emerged in the 1970s as potential solutions for safer, more stable lithium batteries. Initial research focused on PEO-based SPEs, but their poor ionic conductivity hampered performance. Since then, research has expanded to explore a range of polymer matrices, including PAN, PVDF, PMMA, and PC, each with unique properties and challenges (Figure 3) (Jia *et al.*, 2024).

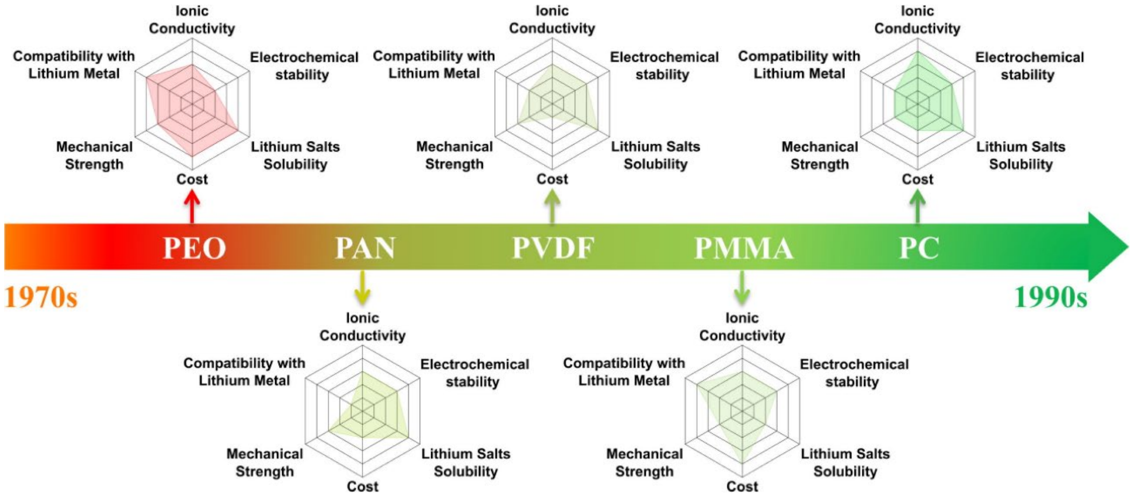


Figure 3 A concise overview of the historical progression and characteristics of diverse polymer matrices (reprinted from (Jia *et al.*, 2024)).

Polymer electrolyte can be classified into a few categories (Figure 4). Solid polymer electrolytes (SPEs) are entirely solid and consist of a polymer matrix with a dissolved salt for ion conduction. Gel polymer electrolytes (GPEs) combine aspects of solid and liquid electrolytes. They have a polymer matrix, a salt, and a liquid solvent or plasticizer to improve ion mobility, resulting in a gel-like consistency. Composite polymer electrolytes (CPEs) are another variation where solid-phase fillers, often ceramic nanoparticles, are added to the polymer matrix and salt combination. These fillers aim to improve various properties of the electrolyte. All three types (SPEs, GPEs, and CPEs) are under development for use in batteries and other electrochemical devices.

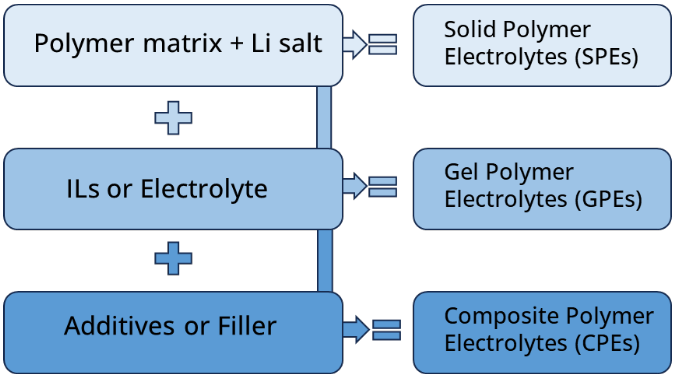


Figure 4 Polymer electrolyte classification based on their composition.

Solid-state energy storage technologies are increasingly sought after for their potential to improve safety, energy density, and the overall sustainability of batteries (Kan *et al.*, 2024; Lu *et al.*, 2024; Zheng *et al.*, 2024). Among these technologies, solid polymer electrolytes (SPEs) and gel polymer electrolytes (GPEs) offer a promising path toward replacing the flammable and potentially toxic liquid electrolytes traditionally used in lithium ion batteries (LIBs) (Baskoro, Wong and Yen, 2019; Sutton *et al.*, 2020). The central component of these electrolytes is the polymer matrix, which critically influences performance characteristics such as ionic conductivity, mechanical stability, and interfacial compatibility with electrodes (X. Ma *et al.*, 2022).

2.4.2 Traditional Polymers in SPEs and GPEs

Several traditional polymers have been extensively explored as the backbone of SPEs and GPEs. The suitability of a polymer as a SPE and GPE hinges on its ability to dissociate lithium salts, yielding mobile Li^+ ions for ionic conduction. This dissociation is facilitated by polar functional groups within the polymer that interact with lithium ions (Chen *et al.*, 2021). Ion transport occurs via a hopping mechanism between coordination sites, with the Arrhenius and Vogel-Tamman-Fulcher (VTF) theories describing the relationship between temperature and conductivity (Zhou *et al.*, 2013). Ideally, the polymer host should have a high dielectric constant and the salt a low lattice energy to facilitate dissociation and promote segmental motion. Importantly, ionic conduction primarily occurs in amorphous polymer regions, making a low glass transition temperature desirable (Al-Akhras *et al.*, 2020). Here's an overview of their key properties, advantages, and the strategies employed to overcome their limitations.

Polyethylene oxide (PEO): PEO remains a popular choice for SPEs due to its cost-effectiveness, mechanical stability, ability to dissolve lithium salts, and compatibility with lithium metal anodes (LMAs) (J. Song *et al.*, 2023). It contains amorphous and crystalline phases, with ionic transport primarily occurring in the amorphous phase (Johari *et al.*, 2021). Oxygen atoms on PEO chains facilitate coordination with and transfer of Li^+. While PEO's low conductivity at room temperature and narrow electrochemical stability window (<3.8 V) pose challenges, various strategies are available (Chen *et al.*, 2023). Modifying the polymer structure and introducing inorganic fillers have proven effective at enhancing both conductivity and stability (X. Li *et al.*, 2023). Crosslinking PEO segments offers another effective approach. Kuo et al. created a cross-linked PEO network with PAN, resulting in high electrolyte uptake, reduced crystallinity, and improved mechanical stability (Kuo *et al.*, 2014). The addition of Poly(dimethylsiloxane) (PDMS) further enhanced electrolyte uptake and lithium ion transference number (Tsao and Kuo, 2015). Physical methods, such as UV-induced crosslinking of a trimethylolpropane ethoxylate triacrylate ionomer (demonstrated by Li et al.), offer an alternative approach, resulting in good ionic conductivity, high lithium ion transference, and excellent thermal and mechanical stability (W. Li *et al.*, 2017). Finally, electron beam radiation, as utilized by Zhang et al. to crosslink core (PAN)-shell (PEO) nanofiber membranes, provides another promising route toward GPEs with high ionic conductivity, electrochemical stability, and appropriate lithium ion transference (Zhang *et al.*, 2015).

Polyacrylonitrile (PAN): PAN offers excellent electrochemical and thermal stability, making it suitable for high-voltage batteries (Yang Wang *et al.*, 2023). Its polar nitrile groups also enable higher ionic conductivity than PEO at room temperature (Zhao and Tenhaeff, 2019). However, PAN-based SPEs suffer from brittleness and poor compatibility with LMAs, potentially leading to increased interfacial resistance (Tian, Kim and Kim, 2024). These issues can be addressed through copolymerization and cross-linking, leading to improved interfacial stability (Nam *et al.*, 2020). Additionally, various modifications have been explored. For example, GPE composite fabricated by immersing PAN/Al_2O_3 electrospun membrane in triethylene glycol diacetate-2-propenoic acid butyl ester (TEGDA-BA), achieving enhanced electrolyte uptake, mechanical properties, and electrode compatibility (Zhu *et al.*, 2019). Yang et al. incorporated octa-(3-chloropropyl)-polyhedral oligomeric silsesquioxane (OCP-POSS) into a PVDF/PAN/PMMA blend, resulting in improved electrolyte uptake, thermal stability, ionic conductivity, and electrochemical stability window (Yang *et al.*, 2017).

Polymethyl methacrylate (PMMA): PMMA offers several advantages: its carbonyl groups promote lithium salt dissociation, its amorphous nature aids ionic conduction, and it has good affinity with LMAs for uniform lithium deposition. Additionally, PMMA is relatively inexpensive (Aadesh *et al.*, 2023). However, mechanical strength is a significant drawback, and PMMA-based SPEs often require plasticizers to improve conductivity, raising flammability concerns (Su'ait *et al.*, 2009). Strategies like blending with high-strength polymers (e.g., PVDF) or incorporating inorganic fillers can mitigate these drawbacks (Zhai *et al.*, 2022). To enhance its stability, strategies like blending, copolymerization, and the introduction of nanofillers are commonly employed. For example, Guan et al. synthesized di-block copolymers of PMMA-block-polystyrene (PS), where the rigid PS block improved mechanical and thermal stability, while the PMMA block maintained favorable ionic conductivity (Guan *et al.*, 2016). Similarly, Huang et al. used a poly(propylene carbonate) (PPC)/PMMA coating on a Celgard separator, resulting in good ionic conductivity, rate capability, and cycling performance(Huang *et al.*, 2017).

Polycarbonates (PC): PCs are appealing due to their high polarity, facilitating salt dissociation, their thermal stability, and oxidation resistance, which are ideal for high-voltage batteries (Motokucho *et al.*, 2018). However, PC-based SPEs suffer from low mechanical strength and poor interfacial compatibility with LMAs (Huo *et al.*, 2023). Approaches like adding support skeletons (cellulose, ceramic) can improve mechanical strength, but the high reactivity between PC and LMAs remains a significant hurdle for the safety and stability of these electrolytes (C. Wang *et al.*, 2018).

Polyvinylidene fluoride (PVDF): PVDF boasts a high dielectric constant, promoting the dissociation of lithium salts and providing a high concentration of charge carriers. Its flexibility, mechanical strength, thermal stability, and wide electrochemical window make it a compelling SPE material (Meng, Lian and Cui, 2021). However, PVDF's high crystallinity limits ionic conductivity, which can be addressed through copolymerization (e.g., with Trifluoro ethylene ;TrFE) or the introduction of inorganic fillers to enhance conductivity by disrupting crystallinity and improving dimensional stability (Fu *et al.*, 2023; Kim *et al.*, 2024).

Poly(vinylidene fluoride-hexafluoropropylene) (PVDF-HFP): A vital component for both GPEs and SPEs, PVDF-HFP is critically important for electrolyte development, offering key advantages. PVDF-HFP exhibits several advantages as an SPE matrix. It possesses a low degree of crystallinity, good thermal and solvent resistance, and a low glass transition temperature (-62 °C) (Xie *et al.*, 2008). The presence of HFP monomers disrupts regular chain arrangements compared to PVDF alone, increasing amorphous regions (Ataollahi *et al.*, 2012). While its fluorine content provides mechanical strength and some hydrophobicity, PVDF-HFP readily dissolves lithium salts due to its polarity (Mariappan *et al.*, 2020). Ionic conductivity in PVDF-HFP depends on the ratio of functional groups to dissociated lithium ions and is influenced by polymer molecular weight (Sangeetha *et al.*, 2021).

Despite these advantages, room-temperature ionic conductivity in PVDF-HFP remains limited (Li *et al.*, 2019). Strategies to address this include blending with other polymers or introducing plasticizers to further increase the amorphous phase (Das and Ghosh, 2017). Research has demonstrated that blending PVDF-HFP with polymers like PEO, PAN, or PMMA can significantly enhance ionic conductivity (Yanılmaz and Zhang, 2015). However, plasticizers can compromise mechanical strength (Halder, Mohamed, S.-W. Kuo, *et al.*, 2024). Alternatively, incorporating fillers (solid plasticizers) into the system offers the potential to improve both ionic conductivity and mechanical robustness, making composite polymer electrolytes a promising avenue for future SPE development (Halder, Mohamed, S.-W. Kuo, *et al.*, 2024).

2.4.3 Green and Biodegradable Options

The search for environmentally conscious energy storage solutions has fuelled the development of biodegradable polymer electrolytes (GPEs). Among natural polymers, cellulose and algal-based polysaccharides offer distinct advantages for GPE applications.

Cellulose, the most abundant natural polymer, is known for its excellent thermal stability and affinity for liquid electrolytes due to its abundant hydroxyl groups (Lizundia and Kundu, 2021). Researchers have explored various forms of cellulose to create GPE hosts. For example, Xiao et al. prepared water-soluble methylcellulose (MC) GPEs demonstrating improved ionic conductivity and lithium ion transference number compared to commercial separators (S. Xiao *et al.*, 2014). For enhanced safety, dense hydroxyethyl cellulose (HEC) membranes offer high-temperature resistance. Zhu's group fabricated porous carboxymethyl cellulose (CMC) membranes to optimize ionic conductivity (Zhu *et al.*, 2015), while Wan et al. created nano-porous cellulose aerogel membranes capable of excellent performance even at high temperatures (Wan *et al.*, 2017).

To mitigate potential issues with residual hydroxyl groups in MC-based GPEs, composite membranes (e.g., PVDF/MC/PVDF) and nonwoven fabrics provide support (S. Y. Xiao *et al.*, 2014; M. Li *et al.*, 2015). Blending cellulose with traditional polymers has yielded positive results (Miri *et al.*, 2015). Combination of thermoplastic polyurethane and cellulose, are able to achieve a high lithium ion transference number and excellent cycling stability (K. Liu *et al.*, 2016). Additionally, blending poly(ethylene glycol methacrylate) (PEGMA) with cellulose triacetate (CTA) addresses CTA's high crystallinity, enhancing both mechanical and ion transport properties (Nirmale *et al.*, 2017). Electrospinning has emerged as a valuable technique for creating porous GPE skeletons, as demonstrated by Kang's group's work with electrospun PVDF/cellulose acetate (CA) non-woven membranes that exhibit enhanced ionic conductivity (Kang *et al.*, 2016).

Beyond cellulose, other bio-based materials like lignin, micro-fibrillated cellulose (MFC), and bacterial cellulose are also being explored for GPEs (Zhu *et al.*, 2019). Zhu et al. demonstrated improved ionic conductivity and cycling performance by incorporating halloysite nanotubes into a cellulose acetate/poly-l-lactic acid (CA/PLLA) blend GPE (Zhu, Lan, *et al.*, 2016). For a greener fabrication process, Zhu et al. created soy protein isolate/polyvinyl alcohol (SPI/PVA) composite nanofiber membranes that effectively leverage SPI's functional groups for efficient lithium ion transport (Zhu, Tan, *et al.*, 2016).

Algal-based polysaccharides offer a compelling and sustainable alternative for GPE development. Agar and agarose, derived from red algae, are versatile materials for creating solid, quasi-solid, gel, and aerogel electrolytes (Kasprzak, Mayorga-Martinez and Pumera, 2023). Researchers have extensively explored ammonium salts (e.g., NH_4SCN, NH_4I, NH_4Br, NH_4NO_3) as additives to agar-based electrolytes to boost proton conductivity – a key feature for fuel cells and proton batteries (Torres and De-la-Torre, 2021). Ethylene carbonate can further increase conductivity by acting as a cosolvent (Torres and De-la-Torre, 2021). Gel electrolytes, formed by trapping liquid electrolytes within an agar matrix, offer high ionic conductivity, long-term stability, and leak resistance (Raphael, Jara and Schiavon, 2017). Researchers have explored salts ($KClO_4$, $LiClO_4$) and weak acids (acetic, lactic) as additives within agar-based gel electrolytes, with $KClO_4$ demonstrating the highest conductivity (Torres and De-la-Torre, 2021). The incorporation of iodide salts (NaI, KI) and the ionic liquid 1-methyl-3-propyl imidazolium (MPII) into agar-based quasi-solid electrolytes has shown promise in enhancing ionic conductivity (Nadia *et al.*, 2017). FTIR analysis suggests complexation between KI and MPII within the polymer matrix, contributing to the conductivity improvement (Ming, Ramesh and Ramesh, 2016).

Bio-based polymers from sources like cellulose and algae provide exciting avenues for developing high-performance, safe, and environmentally responsible GPEs. The variety of available biopolymers, their unique properties, the flexibility to combine them with traditional polymers, and the ability to create advanced structures through techniques like electrospinning offer numerous possibilities for the next generation of sustainable energy storage solutions.

Traditional polymers like PEO, PAN, PVDF, and its derivative PVDF-HFP play crucial roles in SPEs and GPEs. Each polymer offers specific advantages and faces limitations. Strategies like blending, copolymerization, and introducing fillers are employed to improve ionic conductivity, mechanical strength, and interfacial compatibility. With growing environmental concerns, biodegradable polymers derived from sources like cellulose are gaining attention for developing greener electrolytes. While polymer choice is fundamental, achieving the optimal performance of SPEs and GPEs often requires careful engineering using additives and fillers. These materials play a multifaceted role in enhancing ionic conductivity, mechanical robustness, and interfacial stability – factors critical for practical solid-state battery applications.

2.4.4 The Role of Fillers in CPEs

Fillers play a multifaceted role in improving CPEs performance. They are broadly separated into two categories:

- Inactive Fillers: These fillers primarily enhance ionic conductivity through mechanical effects (S. Liu *et al.*, 2023). For example, nanosized materials like Al_2O_3, TiO_2, and SiO_2 can disrupt polymer chain crystallization, increasing the amorphous regions where ion transport is favored. This approach is straightforward but can sometimes compromise mechanical stability (Johari *et al.*, 2021).
- Active Fillers: These fillers participate directly in Li^+ transport. They include materials with Lewis acid-base properties (e.g., ZrO_2, $Li_7La_3Zr2O_{12}$ (LLZO)) and fast ion conductor materials (e.g., $Li_{0.33}La_{0.557}TiO_3$ (LLTO), $Li_{6.4}La_3Zr_{1.4}Ta_{0.6}O_{12}$(LLZTO)). These active fillers create additional conduction pathways within the CPEs (F. Zheng *et al.*, 2023; Z. Li *et al.*, 2023).

Filler characteristics are crucial for maximizing their effectiveness:

- Dimensionality: 1D (nanowires, nanotubes), 2D (nanosheets), and even 3D framework fillers have been explored. 1D and 3D fillers offer a greater potential for building continuous and well-aligned Li^+ conduction pathways, boosting ionic conductivity more effectively than 0D nanoparticles (F. Zheng *et al.*, 2023).
- Size: Filler size profoundly impacts CSPE properties. Nanoscale fillers generally have a larger surface area, increasing the interactions between the filler, polymer, and lithium salt, aiding in salt dissociation and often improving conductivity (Zafar, Shah and Hussain, 2024).
- Surface Properties: Surface functionalization of fillers can significantly enhance CPE performance. For example, coating LLZO nanoparticles with LiF has been shown to improve interfacial stability with lithium metal anodes (Sun *et al.*, 2020).

2.4.5 Strategies and Considerations

While fillers play a critical role, other aspects of CPEs design and optimization include:

- Polymer Matrix Engineering: Polymer selection is fundamental. High polarity polymers like PEO, PVDF-HFP, and PAN promote lithium salt dissociation. Polymer blending, crosslinking, and block copolymer architectures can further enhance properties (Xi *et al.*, 2021).
- Plasticizers: Introducing plasticizers (e.g., succinonitrile, carbonates) increases amorphicity and chain flexibility, improving ionic conductivity but potentially sacrificing mechanical strength (Yue *et al.*, 2016).
- Functionalization: Functionalizing polymers and fillers can promote specific interactions with lithium ions or anions, helping to increase ion mobility and lithium ion transference number (Chen *et al.*, 2021).
- Thin and Robust CPEs: Achieving CPEs with the thinness of commercial separators (around 20 μm) while maintaining mechanical strength is crucial for practical SSBs. Techniques like electrospinning and layer-by-layer assembly hold promise in this area (Wang *et al.*, 2024).

2.4.6 Future Outlook: Challenges and Opportunities

Despite tremendous progress, several challenges in CPE development remain:

- Room-Temperature Conductivity: Further increases in ionic conductivity exceeding 10^{-2} S cm^{-1} are needed to rival liquid electrolytes (Yuan *et al.*, 2022).

- Lithium ion Transference Number: Approaches for selectively immobilizing anions will be key to increasing the Li^+ transference number (Jia, Khurram Tufail and Guo, 2023).
- Scalability: Transitioning CPEs fabrication from the lab to large-scale manufacturing at a reasonable cost is essential (Fan, He and Nan, 2021).
- Thermal Stability and Safety**:** Incorporating flame-retardant elements or additives will be crucial for safe SSB operation (Q. Wang *et al.*, 2021).

Overall, the continued development of CPEs, particularly through strategic filler design, functionalization, and optimization of CPE composition, promises to unlock the full potential of polymer electrolyte. These efforts will be central to creating safer, higher-energy density, and more sustainable energy storage solutions.

2.4.7 Polymer Electrolyte Fabrication Method

Several methods exist for preparing polymer electrolytes, each with its advantages and considerations. Let's delve into the most common techniques:

2.4.7.1 Solution Casting

Solution casting is a straightforward and versatile approach favoured for its simplicity. In this method, the polymer matrix and salt electrolytes are dissolved in a suitable solvent to create a homogeneous solution. This solution is then cast onto a substrate and dried at a low temperature to evaporate the solvent (J. Gao *et al.*, 2021). Low-boiling-point solvents are preferred to facilitate easy removal. The advantage of this method lies in its adaptability; GPE properties can be tuned by adjusting the weight and thickness of the cast layer. Solution-cast GPEs are flexible, pressure-resistant, and non-volatile. However, their low crystallinity may lead to reduced mechanical strength, which can be mitigated by introducing crosslinking agents to enhance structural integrity (Montanheiro *et al.*, 2021). Since moisture can interfere with electrolyte performance, careful handling is crucial in this process.

For polymers like PVDF and PVDF-HFP, high-boiling-point solvents are necessary. In these cases, plasticizers (e.g., carbonates, polyethylene glycol) or pore-forming agents are often incorporated to enhance polymer chain mobility and create a porous structure upon removal. The GPE is then activated by immersion in a liquid electrolyte, allowing it to fill the voids and enhance conductivity. Thorough removal of solvents and additives is vital for a strong and stable GPE (Aruchamy *et al.*, 2023).

2.4.7.2 Phase Inversion

The phase inversion method offers a scalable and cost-effective way to produce GPEs (Huang *et al.*, 2015). Here, a polymer solution or polymer-liquid composite is solidified by introducing a non-solvent. The polymer solution is coated onto a substrate and then immersed in the non-solvent, which replaces the original solvent and causes the polymer to precipitate (Amaral *et al.*, 2015). This process can be tuned to create honeycomb-like or network morphologies within the GPE (Ghahramani *et al.*, 2023), with the latter being ideal for efficient ion transport and reduced electrolyte leakage. Phase inversion allows for continuous, defect-free GPEs but requires careful optimization of solvent, non-solvent, and polymer concentration (Montanheiro *et al.*, 2021).

2.4.7.3 In Situ Polymerization

In this method, polymerization of the matrix occurs directly within the liquid electrolyte. Monomers or prepolymers, initiators, and the liquid electrolyte are mixed and then subjected to heat or light to trigger polymerization (C. Ma *et al.*, 2022). This results in a GPE with an uninterrupted ion transport network. Crucially, since the mixture can be coated onto an electrode before polymerization, in situ methods can lead to excellent electrode-electrolyte interfaces, reducing interfacial resistance and potentially hindering lithium dendrite growth (Vijayakumar *et al.*, 2021). Advantages include low viscosity mixtures for ease of handling and seamless interface formation (Cui *et al.*, 2017). However, the materials needed can be expensive, and thermal stability is often lower compared to bulk polymers like PVDF, PEO, and PVA (Aruchamy *et al.*, 2023).

2.4.7.4 Electrospinning

Electrospinning transforms a polymer solution into a porous layer of nanofibers. A high voltage is applied to the solution, drawing it into fibers that are collected on a conductive substrate (Xue *et al.*, 2019). The thickness of this layer is controlled by the solution volume, while factors like concentration, voltage, and solvent choice influence fiber thickness (Chenyu Wang, Wang, *et al.*, 2019). This fibrous matrix then serves as the GPE scaffold, with liquid electrolyte impregnated into the interconnected pores. Electrospinning offers a larger surface area for conduction and reduces solvent usage. However, high-viscosity polymer solutions are required, and when working with polymers like PVDF, non-volatile solvents necessitate careful handling for effective drying (Chang and Lai, 2020).

2.4.7.5 3D printing

3D printing with bio-based polymers offers a unique and sustainable approach to crafting energy conversion and storage devices (Firmanda *et al.*, 2022). This method's geometric flexibility enables optimized structures, while bio-based polymers provide printability, cost-effectiveness, biocompatibility, and environmental benefits. This combination has the potential to revolutionize applications such as implantable energy storage, wearable electronics, and bio-integrated systems (Putri, Intasanta and Hoven, 2024). The technology aligns with the UN's Sustainable Development Goals, promoting responsible consumption and clean energy(Putri, Intasanta and Hoven, 2024). While extrusion-based printing currently dominates, its limitations include porosity control and technology cost (Chen *et al.*, 2020; Yao *et al.*, 2021). Advancements in direct ink writing, high-resolution nozzles, and ink development will enhance the performance of 3D-printed devices (Park *et al.*, 2022; Saadi *et al.*, 2022). Alternative techniques, like stereolithography, offer advantages in intricate structure creation (Cheng, Deivanayagam and Shahbazian-Yassar, 2020). Exploring underutilized bio-based polymers like chitin and chitosan could reveal new possibilities for conductivity and charge transfer (Peter *et al.*, 2021; Anwer *et al.*, 2023). Overall, the integration of 3D printing with bio-based polymers promises advancements in sustainable energy solutions, contributing to a greener future.

2.4.8 Summary

This literature review section delves into the current state of polymer electrolyte research. It investigates traditional polymer matrices, their advantages and limitations, and explores the use of additives and fillers to enhance electrolyte performance. Definitions of different polymer electrolyte types are provided, alongside a survey of fabrication methods, both established and emerging. This collected knowledge will be instrumental in guiding the design of future polymer electrolytes.

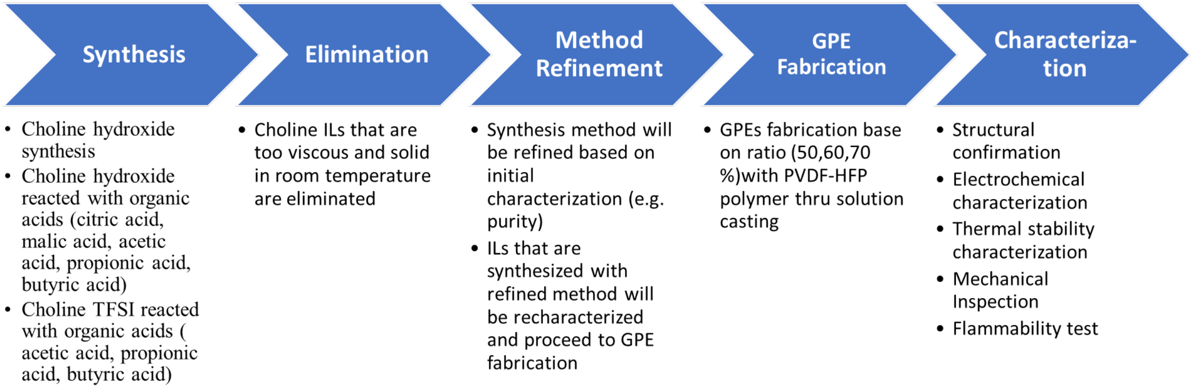
2.5 Overall Summary

This section of literature review reveals the complex landscape of electrolyte technologies for lithium ion batteries. While a range of options exist (organic, aqueous, GPEs, solid-state), each possesses inherent limitations and advantages. Challenges such as safety concerns with organic electrolytes, electrolyte leakage, thermal runaway, and limited electrochemical stability windows persist across these categories.

This section also highlights the potential of ionic liquids (ILs), deep eutectic solvents (DESs), and polymeric ionic liquids (PILs) as alternatives. Choline-based ILs emerge as particularly promising candidates due to their affordability, broad electrochemical potential windows, and versatile applications in energy devices like batteries, supercapacitors, and solar cells. Within this domain, polymer electrolytes offer a path toward addressing leakage, safety, and mechanical integrity concerns. Traditional polymer matrices and emerging green/biodegradable options show promise, while the use of fillers in composite polymer electrolytes (CPEs) introduces opportunities for performance optimization.

Chapter 3: Materials and Methods

3.1 Experiment Flow Diagram



Synthesis

- Choline hydroxide synthesis
- Choline hydroxide reacted with organic acids (citric acid, malic acid, acetic acid, propionic acid, butyric acid)
- Choline TFSI reacted with organic acids (acetic acid, propionic acid, butyric acid)

Elimination

- Choline ILs that are too viscous and solid in room temperature are eliminated

Method Refinement

- Synthesis method will be refined based on initial characterization (e.g. purity)
- ILs that are synthesized with refined method will be recharacterized and proceed to GPE fabrication

GPE Fabrication

- GPEs fabrication base on ratio (50,60,70 %)with PVDF-HFP polymer thru solution casting

Characteriza-tion

- Structural confirmation
- Electrochemical characterization
- Thermal stability characterization
- Mechanical Inspection
- Flammability test

3.2 Chemicals and Materials

Choline chloride was obtained from Acros. Citric acid from Fisher Scientific. Choline bis(trifluoromethane)sulfonimide (ChTFSI) was obtained from Lolitec. Ethanoic acid, acetone, and ethyl acetate was obtained from EMSURE, propanoic acid and butanoic acid were acquired from R&M, Lithium bis(trifluoromethanesulfonyl)imide (LiTFSI), dimethyl sulfoxide (DMSO), concentrated sulphuric acid, sodium bicarbonate, malic acid, potassium hydroxide and poly(vinylidene fluoride)-co-hexafluoropropylene (PVDF-HFP) were acquired from Sigma Aldrich. All chemicals used are analytical grade and used without further purification. All the battery-grade materials for battery fabrication were acquired from KCG Resources Sdn Bhd, which includes lithium cobalt oxide (LCO), Polyvinylidene fluoride (PVDF), Super P (conductive carbon), graphite, CR2032 coin cell casing, aluminium foil, and copper foil.

3.3 Synthesis of Choline Hydroxide

Choline hydroxide was synthesized through an acid-base reaction. Choline chloride was dissolved in ethanol in a conical flask, followed by the addition of potassium hydroxide in a 1:1 molar ratio. The mixture of choline chloride, potassium hydroxide, and ethanol was stirred for 12 hours at 60°C under atmospheric pressure within a fume hood. This process resulted in the formation of choline hydroxide (ChOH) and a precipitate of potassium chloride (KCl) within the conical flask. The KCl precipitate was then removed by filtration using a vacuum filter. The filtered product contained choline hydroxide with excess cooled ethanol. Subsequently, ethanol was separated from the product using a rotary evaporator with a water bath set at 80°C. During this step, any remaining KCl that was unable to be filtered out earlier crystallized. Another vacuum filtration process was performed to remove the remaining KCl. In the synthesis of choline hydroxide, potassium chloride (KCl) is formed as a precipitate due to the acid-base reaction between choline chloride and potassium hydroxide. This reaction produces choline hydroxide and KCl as by-products:

$$\text{Choline chloride} + \text{KOH} \rightarrow \text{Choline hydroxide} + \text{KCl}$$

KCl has limited solubility in ethanol (the solvent used in this reaction), and as the reaction proceeds, the concentration of KCl exceeds its solubility limit, causing it to precipitate out of the solution. The solid KCl is then removed by vacuum filtration. Any residual KCl that remains dissolved can recrystallize during the ethanol evaporation step, making it easy to filter out completely.

Finally, a viscous and yellowish choline hydroxide was obtained as the product. This method was adapted from the work of Vajekar and Shankarling (Vajekar and Shankarling, 2020). The process of the synthesis is illustrated in Figure 5.

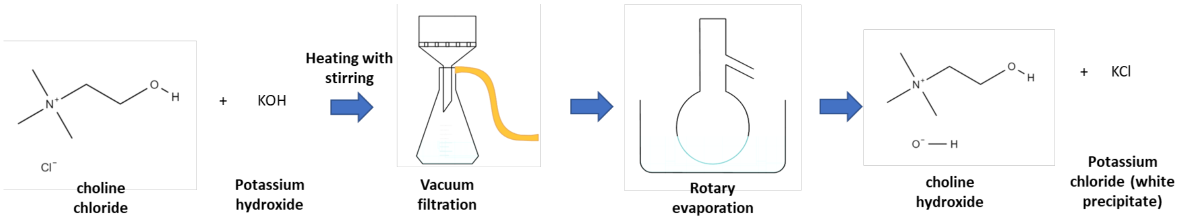


Figure 5. schematic diagram of the synthesis of choline hydroxide.

3.4 Synthesis of Choline Based Ionic Liquid With Choline Hydroxide.

Choline hydroxide and the corresponding organic acids (malic acid, citric acid, acetic acid, propionic acid, and butyric acid) were added to a conical flask in a 1:1 molar ratio. The mixture was stirred for 12 hours at room temperature and atmospheric pressure, producing a yellowish and viscous solvent. To remove unreacted acid, ethyl acetate was added to the conical flask. This addition led to forming two distinct layers, one clear and colourless, and the other cloudy, yellowish, and viscous, settled at the bottom of the flask. The clear, colourless layer was identified as water, while the yellowish layer was identified as the DESs. The mixture was then transferred to a separation funnel and allowed to settle for one hour to facilitate the separation of the ionic liquid layer. The DES layer was separated from the funnel and transferred to a vacuum oven for drying. The vacuum oven was set to 25°C and maintained under vacuum pressure for 3 days to complete the drying process. (Quental *et al.*, 2015). The process of the synthesis is illustrated in Figure 6.

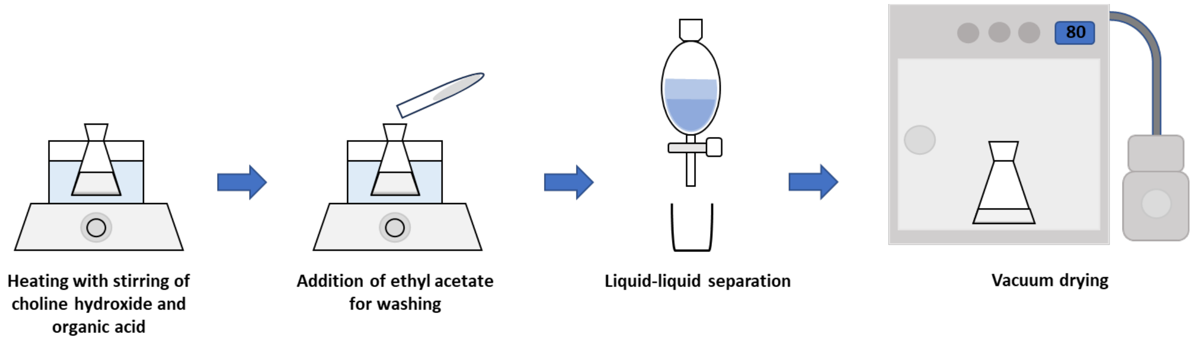


Figure 6. Schematic diagram of the synthesis of choline-based ionic liquid.

3.5 Synthesis Choline based Ionic Liquids with Choline TFSI.

The synthesis of choline-based ionic liquids with choline TFSI involved an esterification reaction where choline reacted with a carboxylic acid such as acetic acid, propionic acid, and butyric acid. This reaction produces the desired choline ester ionic liquid and water as a byproduct in a reversible reaction (Equation 1). The detailed procedure is described in the following section, where methods are first used and after refinement.

$$\underset{\text{(choline TFSI)}}{[\,(CH_3)_3NCH_2CH_2OH]^+\,[TFSI]^-} + \underset{\textit{(organic acid)}}{X\text{-}COOH} \xrightleftharpoons{\textit{Conc. } H_2SO_4 \textit{ (Catalyst)}} \underset{\textit{(choline ester)}}{[X\text{-}COOCH_2CH_2N(CH_3)_3]^+\,[TFSI\,]^-} + \underset{\textit{(water)}}{H_2O} \qquad (1)$$

$X = CH_3,\ CH_2CH_3,\ CH_2CH_2CH_3$

3.5.1 Method Before Refinement

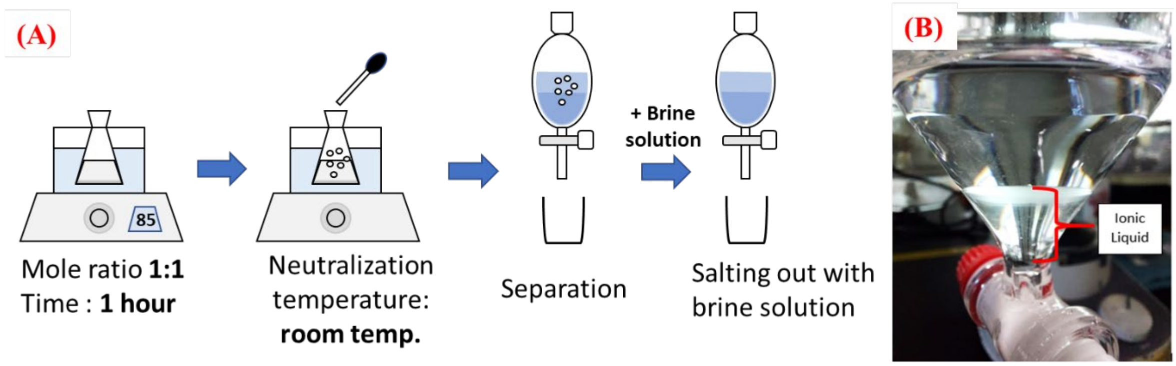


Figure 7. (A) Schematic flow diagram of the synthesis of choline-based ionic liquid choline TFSI before refinement, and photograph of the biphasic separation of the ionic liquid and water (B)

Equal molar amounts of choline TFSI and the selected carboxylic acid were combined and heated with stirring for 45 minutes. A few drops of concentrated sulfuric acid were added as a catalyst. A two-phase mixture was observed at the end of the reaction, indicating the formation of the hydrophobic ester. Excess unreacted carboxylic acids were neutralized by the addition of concentrated sodium bicarbonate. Neutralization was considered complete when no further gas evolution (bubbles) was observed. The mixture was then separated using a separating funnel (Figure 7B). After separation, the ionic liquid phase was washed with ultrapure water followed by brine (to remove residual water). The three newly synthesized ionic liquids were abbreviated based on the specific carboxylic acid used.

3.5.2 Method after refinement

The synthesis of choline-based ionic liquid with choline TFSI was refined is based on a refined method as described in Figure 8.

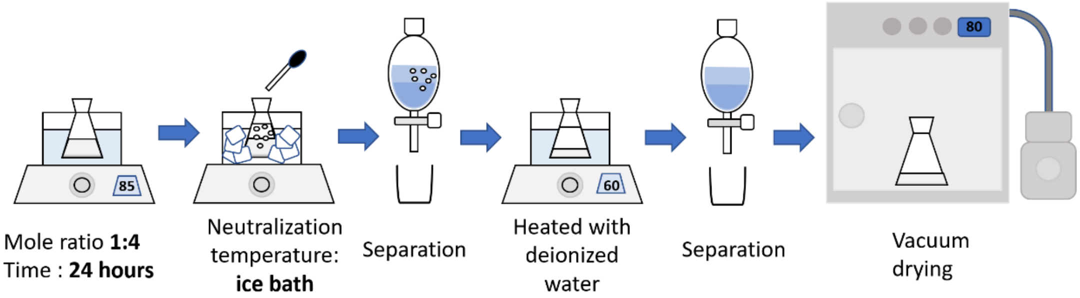


Figure 8. Schematic flow diagram of the synthesis of choline-based ionic liquid with choline TFSI after refinement

The mole ratio between choline TFSI and the carboxylic acid is set to 1:4, which allows the reaction equilibrium to push in the favourable direction. Sulphuric acid was used as the catalyst in the reaction. The reaction was allowed to react for 24 hours with vigorous stirring. After the reaction was completed, it was transferred into an ice bath to cool down the mixture. Once the mixture is cooled, a pre-cooled sodium carbonate solution is added dropwise to neutralize the mixture until no gas evolution is observed. A separating funnel was used to separate the end mixture into waste and ionic liquids. The ionic liquids were then purified under heating with deionized water and another separation with a separating funnel followed by vacuum drying and stored in a desiccator.

The procedures described above, while somewhat tedious, can be optimized for economic feasibility on a larger scale. For instance, the synthesis of choline hydroxide involves relatively inexpensive and readily available chemicals like choline chloride and potassium hydroxide. The reaction takes place under mild conditions (atmospheric pressure, 60°C), reducing the energy costs associated with high-temperature or high-pressure processes. However, the 12-hour stirring time and multi-step purification using filtration and rotary evaporation increase the overall time and energy required, which could pose challenges for large-scale production.

To improve economic feasibility, these synthesis could be streamlined by reducing stirring time, using continuous flow reactors, or integrating filtration and evaporation into a more automated system. Additionally, solvents like ethanol can be recovered and reused, minimizing material costs. Scaling up the reaction to industrial levels can also lead to cost efficiencies due to economies of scale.

The choice of certain experimental parameters, such as the molar ratios and reaction conditions, is based on previous studies and theoretical considerations. The 1:1 molar ratio between choline chloride and potassium hydroxide in the synthesis of choline hydroxide ensures complete conversion of the choline chloride into choline hydroxide, avoiding excess by-products. This ratio balances the reactivity of the components while ensuring that the desired product is formed without requiring excess reagent that would complicate purification.

In the refined method for synthesizing choline-based ionic liquids (choline TFSI), the 1:4 mole ratio between choline TFSI and the carboxylic acid helps to shift the equilibrium of the esterification reaction towards the product side, ensuring a higher yield of the desired ionic liquid. This excess of carboxylic acid drives the reaction to completion, making it more efficient. Additionally, the use of sulfuric acid as a catalyst accelerates the reaction, allowing the synthesis to be completed within 24 hours under controlled conditions.

The 12-hour and 24-hour stirring times used in the synthesis, while necessary to ensure complete reaction, may not be economically viable for large-scale production. Long reaction times could result in increased energy consumption and lower throughput in an industrial setting. One possible solution is to investigate the use of mechanical or ultrasonic stirring to reduce the reaction time, or the implementation of continuous flow synthesis, which could maintain the same level of product yield and quality while significantly decreasing the time needed for each batch.

For example, in continuous flow systems, reactions are run in a steady state, reducing reaction time and energy costs while also allowing for easier scale-up. Further optimization studies may help in determining the minimum effective stirring time without compromising the yield or quality of the ionic liquids.

3.6 Reaction Equations Involved

Here are the key reaction equations involved in the synthesis processes:

1. Synthesis of Choline Hydroxide:

$[(CH_3)_3NCH_2CH_2OH]^+Cl^-$ (*Choline chloride*) + KOH → $[(CH_3)_3NCH_2CH_2OH]^+OH^-$ (*Choline hydroxide*) + KCl

2. Synthesis of Choline-based Ionic Liquid with Organic Acids:

$[(CH_3)_3NCH_2CH_2OH]^+OH^-$ (*Choline hydroxide*) + Organic acid → Choline-based ionic liquid + Water

Example with acetic acid:

$[(CH_3)_3NCH_2CH_2OH]^+OH^-$ (*Choline hydroxide*)+ CH_3COOH → $[(CH_3)_3NCH_2CH_2OH]^+CH_3COO^-$ (Choline acetate) + H_2O

3. Synthesis of Choline TFSI-based Ionic Liquid (Esterification):

H_2SO_4 (catalyst)

Choline TFSI + Carboxylic acid → Choline ester + Water

Example with butyric acid:

H_2SO_4 (catalyst)

Choline TFSI + C_3H_7COOH → Choline butyrate + H_2O

These reactions illustrate the formation of the desired choline-based ionic liquids along with by-products like water and KCl, which are efficiently removed during the purification steps.

3.7 Preparation of Gel Polymeric Electrolyte (GPE)

Figure 9 shows a flow diagram of how GPE was fabricated. First, a solution of PVDF-HFP was prepared with 15 g of PVDF-HFP dissolved in 100 mL of DMSO. Then LiTFSI was dissolved in ionic liquid with a weight ratio of 20% w/w. The ionic liquid and lithium salt mixture was mixed with different weight ratios (Table 5) of PVDF-HFP solution. The final mixture is then cast onto a petri dish and dried in a vacuum oven at 70 °C for 24 hours. Once the GPE was dried, it was punched into a circular film with a diameter of 19 mm. Figure 10E shows a photograph of the fabricated free-standing GPE with different compositions of choline-based ionic liquids. Compared to the commercial battery separator polypropylene (opaque white), the fabricated GPE is transparent and flexible.

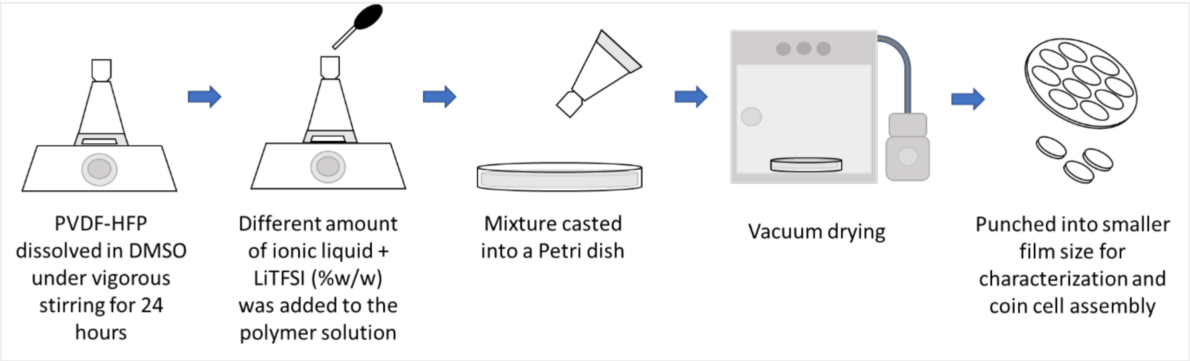


Figure 9. Schematic diagram of the fabrication of ionic liquid gel polymeric electrolyte

Table 5. Weight ratio of ionic liquid, LiTFSI, and PVDF-HFP for GPE fabrication.

Mixture of choline ester and LiTFSI with a ratio of 80:20 (C1)	**Polymer used (C2)**	**Weight ratio C1:C2 (%w/w)**
acetyl choline TFSI + LiTFSI	PVDF-HFP	50:50 60:40 70:30
propionyl choline TFSI + LiTFSI		50:50 60:40 70:30
butyryl choline TFSI + LiTFSI		50:50 60:40 70:30

3.8 Electrode Fabrication and Coin Cell Assembly

The cathode electrode used in this study was fabricated using a mixture of lithium cobalt oxide (LCO), Super P, and polyvinylidene fluoride (PVDF) in a weight ratio of 90:5:5. LCO served as the active material, while Super P improved the electrode's conductivity, and PVDF acted as a binder to hold the material and adhere it to the current collector. The mixture was dissolved in a warmed dimethyl sulfoxide (DMSO) solvent to ensure the complete dissolution of PVDF. The resulting slurry was then cast onto a thin aluminium current collector sheet and spread evenly across the surface using a doctor blade (as illustrated in Figure 10A). The casted film was then dried in a vacuum oven to remove the solvent and punched into circular shapes with a diameter of 16 mm. All the punched electrodes were dried again in a vacuum oven and stored in a glove box.

On the other hand, the anode was fabricated using a technique similar to the cathode but with the mixture having a mix ratio of 96:2:2 (Graphite: PVDF: Super P) on copper foil. These fabricated cathodes and anodes (Figure 10B and C) were then used to assemble a full lithium ion coin cell battery and to be used to characterize the ionic liquid GPEs. The configuration of the assembled coin cell is illustrated in Figure 10D.

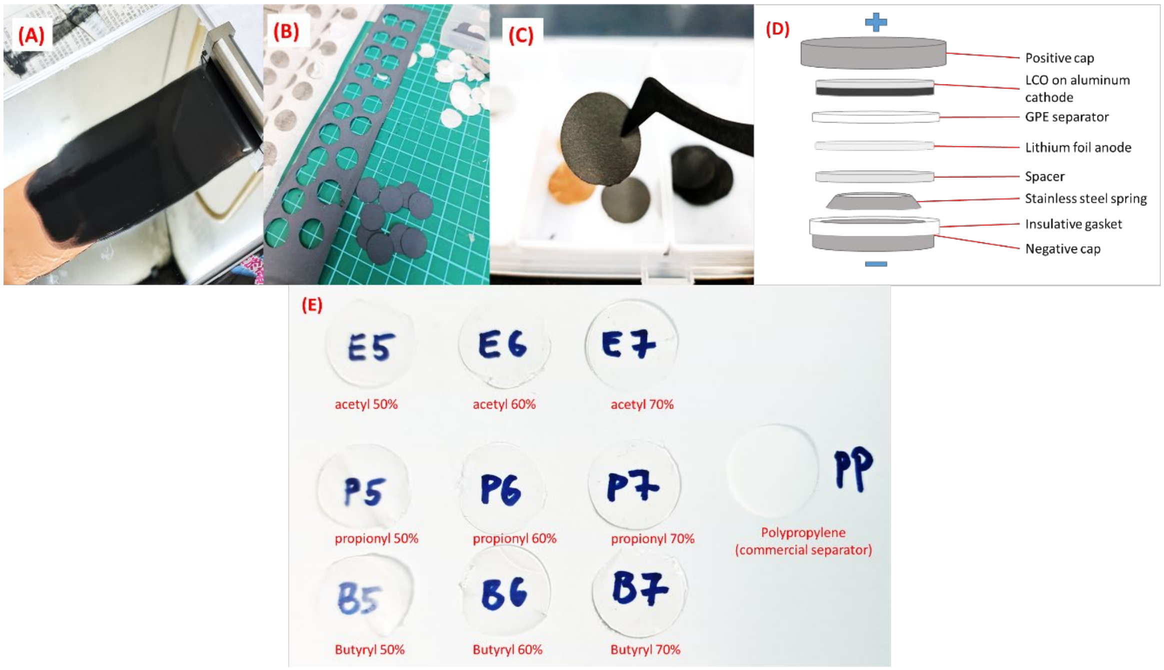


Figure 10. Photograph of (A) casting of electrode material on current collector,(B) dried electrode punched into circular shape, (C) ready-to-use vacuum oven-dried electrode, (D) schematic diagram of coin cell configuration of LCO/Li coin cell, and (E) fabricated ILGPEs with different weight ratio compared with commercial separator.

3.9 Characterization of The Synthesized ILs and GPEs

3.9.1 Viscosity Measurement.

The spindle of the viscometer (Brookfield DV-E viscometer) was set at s61, and the rotational speed was set to obtain results above 20% in percentage torque (%).

3.9.2 Thermal Stability

The thermal properties of the ionic liquids were analyzed using a thermogravimetric analyzer (TGA) and Differential Scanning Calorimeter (DSC) (Mettler Toledo TGA/DSC1). The temperature ranged from 30 to 900 ^{o}C under a nitrogen atmosphere at a heating rate of 10 ^{o}C/min.

3.9.3 Refractive Index Measurement.

The refractive index of the prepared DESs was measured using Optical Refractometer, Abbe 5.

3.9.4 Structural Confirmation

The structure of the synthesized ionic liquid was characterized using Fourier-transform infrared spectroscopy (FTIR)（PerkinElmer Spectrum 3）from 400-4000 cm^{-1} and Nuclear Magnetic Resonance (NMR) (Bruker Ultrashield 300) with ^{1}H NMR, ^{13}C NMR.

3.9.5 Flammability test

A simple flammability test was conducted to demonstrate the ability of prepared GPE towards the fire and compared it to a commercial battery separator (polypropylene).

3.9.6 Electrochemical Properties

Electrochemical evaluation was done to evaluate the feasibility of the synthesized ionic liquid's capability to act as an electrolyte for a lithium battery.

Linear sweep voltammetry (LSV) was used to determine the electrochemical stability window of the electrolytes. The ionic liquids containing LiTFSI were sandwiched between Li metal foil (counter and reference electrode) and stainless-steel foil (working electrode) and scanned between potential from 2-7 V with a scan rate of 0.1 mV/s, the cell configuration is Li ‖ GPE ‖ Stainless Steel.

Moreover, the prepared GPE's ionic conductivities were evaluated using electrochemical impedance spectroscopy (EIS) by applying an alternative voltage of 10 mV over a frequency ranging from 0.01 to 10^5 Hz. The electrical conductivity of the gel polymer electrolyte film is calculated by:

$$\sigma = \frac{l}{R_b A} \tag{2}$$

Where l is the thickness of the polymer electrolyte film, A is the electrode area, and R_b is the bulk resistance. LSV and EIS were measured using an electrochemical workstation (VersaSTAT 3 AMETEK), the cell configuration is Li ‖ GPE ‖ Li.

The lithium ion transference number (t_{Li^+}) was obtained by combining potentiostatic dc-polarisation with ac impedance on the electrochemical workstation system. The Li/electrolyte/Li symmetric CR2032 coin cell for testing was assembled in the glove box. The current of the tested coin cell was recorded with a polarisation voltage of 10 mV until it reached a steady state. The t_{Li}^{+} can be obtained from the formula :

$$t_{Li^+} = \frac{I_{ss}(\Delta\mathrm{V} - I_0 R_0)}{I_0(\Delta\mathrm{V} - I_{ss} R_{ss})} \quad (3)$$

Here, ΔV with a value of 10 mV is expressed for the dc polarisation voltage; R_0 and R_{ss} refer to the interfacial resistance between the electrolyte membrane and the electrodes before and after polarisation; I_0 and I_{ss} represent the initial and steady current, respectively, the cell configuration is Li ‖ GPE ‖ Li.

The fabricated GPEs are also assessed with galvanostatic charge-discharge, which was evaluated using a LAND battery testing system (LANHE CT2001). It was evaluated using a coin cell setting with an LCO cathode and lithium as the anode. The cell was charged and discharged at a constant current of 0.1 mA in the 2.7-4.4 V range, the cell configuration is Li ‖ GPE ‖ Graphite.

Chapter 4: Results and Discussion

4.1 Structural Characterization of Choline Hydroxide

FTIR spectroscopy plays a crucial role in characterizing ILs by providing information about interactions and complexation between constituents, it is also used in some research to investigate the synthesis procedure and interpret the binding mechanism of ILs (Wei *et al.*, 2021). Overall, FTIR spectroscopy is widely used for the structural and compositional analysis of ILs and their interactions with other compounds. FTIR (Fourier Transform Infrared Spectroscopy) is a powerful tool for identifying functional groups and understanding the molecular interactions within a compound. However, it is largely qualitative and provides limited information about the exact structure of a compound. FTIR mainly helps in confirming the presence or absence of specific functional groups by analyzing the characteristic absorption bands. For example, in the case of choline-based ionic liquids, FTIR can be used to identify key functional groups like hydroxyl, carbonyl, or ester linkages, confirming whether a desired chemical reaction has occurred.

However, FTIR alone is not sufficient to definitively determine the full molecular structure of the compound. The peaks in an FTIR spectrum can sometimes overlap, leading to ambiguous results. Therefore, to fully characterize and confirm the structure of the synthesized choline-based ionic liquids, it is crucial to complement FTIR with other techniques, such as: NMR (Nuclear Magnetic Resonance) Spectroscopy, NMR can provide detailed information about the molecular structure, including the arrangement of atoms and the connectivity of functional groups. This is especially useful for confirming the formation of new bonds and the overall structure of the synthesized ionic liquids.

In Figure 11 we can notice that the broad absorption peak around 3264 cm^{-1} in choline hydroxide is a characteristic of O–H stretching vibration. The aliphatic C–H stretching vibration was observed around 2978 cm^{-1}. The methyl bending vibration was observed at 1474 cm^{-1}. The band near 1372 cm^{-1} corresponds to the stretching vibration of the C–N bond. The band at 1087 cm^{-1} corresponds to the C–O stretch in a primary alcoholic group. The C–H bending vibrations of the –CH2– group were observed near 951 and 893 cm^{-1}. Pure choline chloride shows several functional groups, mainly a broad band at 3222 cm^{-1}, associated with the O–H stretching vibration, and a vibration band at 3024 cm^{-1}, which may be attributed to the stretching mode of C–H vibration, was detected. Furthermore, vibration bands at 1480 cm^{-1} were also observed, indicating the presence of an alkyl group and confirming the distinctive properties of choline chloride. Both of the spectra were compared with literature results (Sivrikaya, 2019; Ling *et al.*, 2020; Vajekar and Shankarling, 2020; Yang *et al.*, 2021; Rahmalia *et al.*, 2022), which infers that the choline hydroxide has successfully synthesized and ready to be used for DES preparation with organic acids.

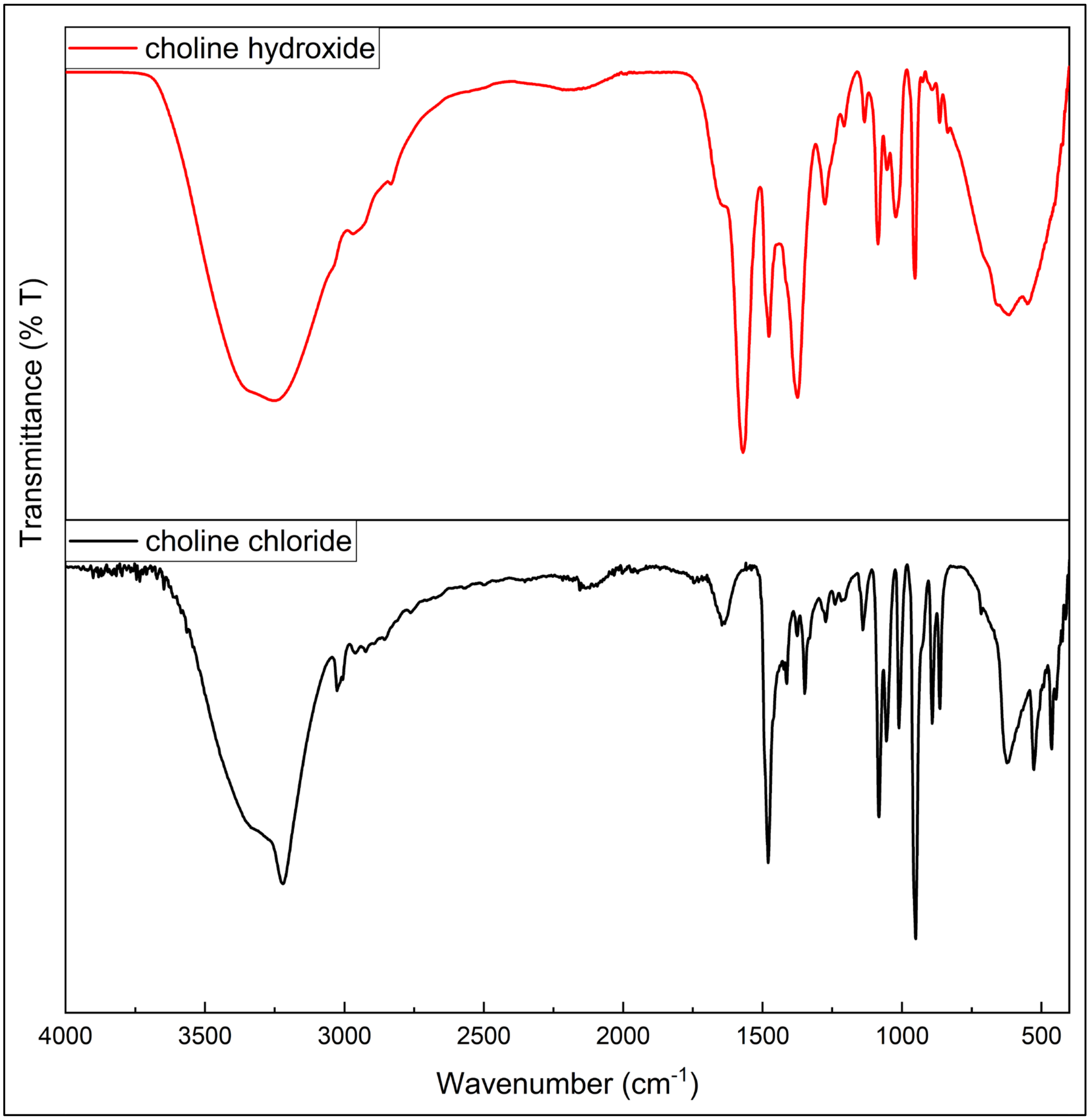


Figure 11. FITR spectra of choline hydroxide and choline chloride.

4.2 Structural Characterization of Choline ILs from Choline Hydroxide

The FTIR spectrum of all the prepared ILs are shown in Figure 12, we can notice that all the ILs show similar absorption band at 3421 cm^{-1} which attributed from the O-H stretching band from the choline hydroxide and organic acids. Notice that then O-H stretching of ILs undergoes a shift to 3421 cm^{-1}, suggesting the presence of hydrogen bonds between organic acid and choline hydroxide during the preparation of ILs (Zullaikah *et al.*, 2018; Sivrikaya, 2019). Also, the C=O stretching band at 1717 cm^{-1} in all the ILs spectrum are characteristic band from the organic acids.

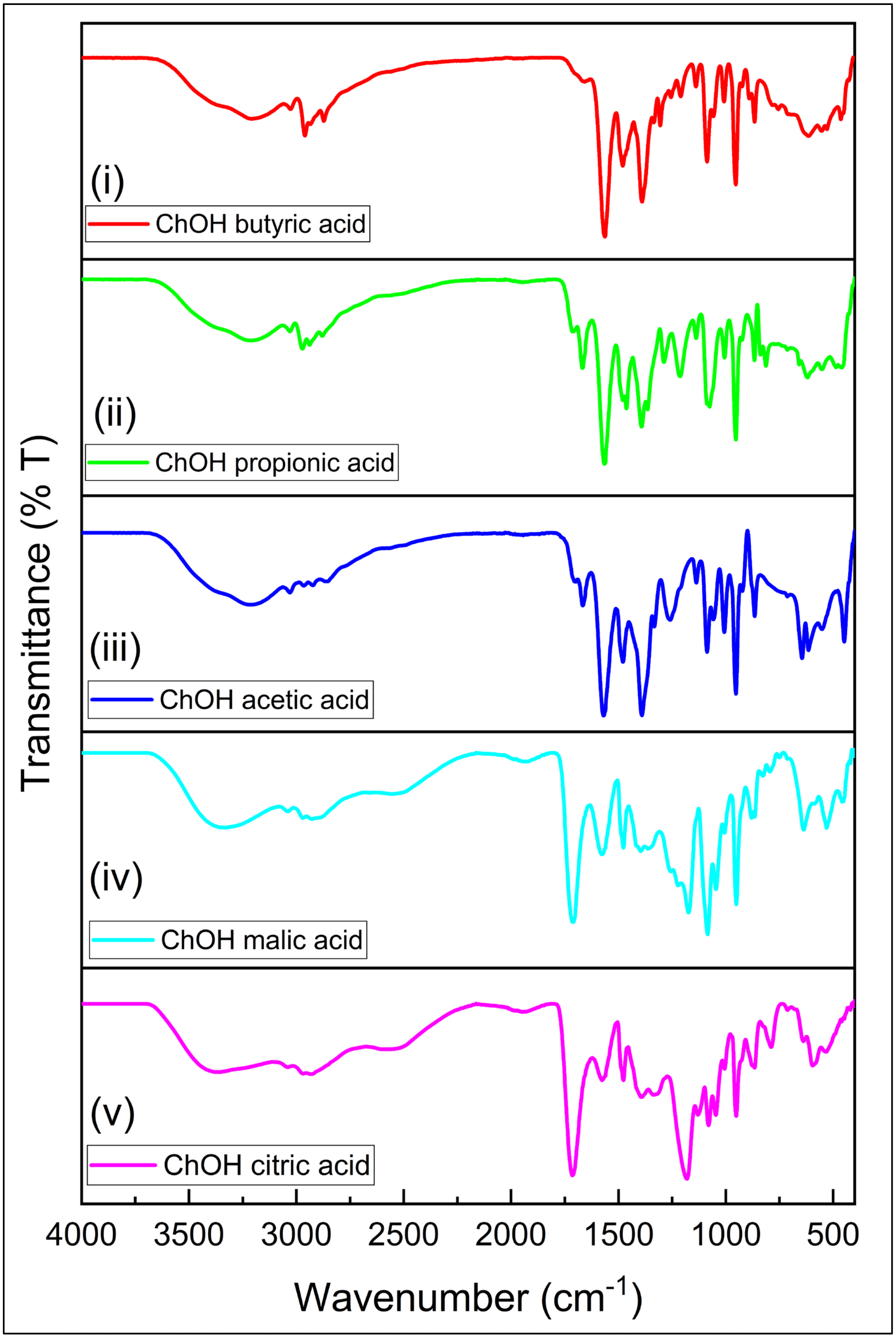

(i)
ChOH butyric acid
(ii)
ChOH propionic acid
(iii)
ChOH acetic acid
(iv)
ChOH malic acid
(v)
ChOH citric acid
Transmittance (% T)
4000
3500
3000
2500
2000
1500
1000
500
Wavenumber (cm-1)

Figure 12. FITR spectra of prepared ILs (i) ChOH butyric acid, (ii) ChOH propionic acid, (iii) ChOH acetic acid, (iv) ChOH malic acid, and (v) ChOH citric acid.

4.3 Thermal Stability of Choline ILs with Choline Hydroxide

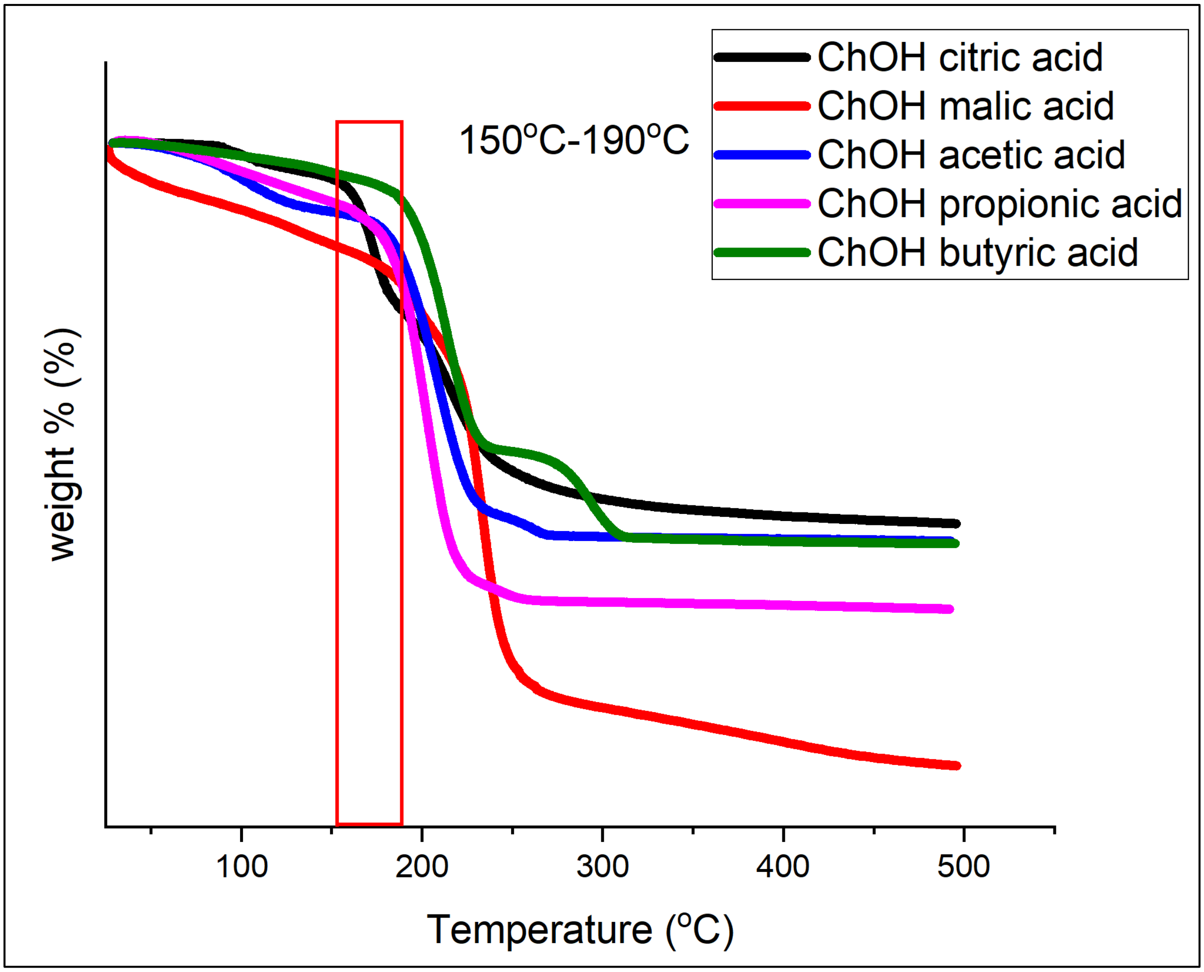


Figure 13. TGA thermogram of prepared DESs ChOH butyric acid, ChOH propionic acid, ChOH acetic acid, ChOH malic acid, and ChOH citric acid.

Thermogravimetric analysis (TGA) is a crucial technique for characterizing the thermal stability and decomposition behavior of ILs (Nuutinen *et al.*, 2019; Ibrahim *et al.*, 2020; Hansen *et al.*, 2021; S. Liu *et al.*, 2022). TGA provides essential data on weight loss and thermal degradation, offering valuable insights into the thermal properties and stability of ILs, which is vital for various applications (Nuutinen *et al.*, 2019; Ibrahim *et al.*, 2020; S. Liu *et al.*, 2022). In this study, TGA was used to investigate the thermostability of the prepared ILs synthesized from choline hydroxide and organic acids; the results are described in Figure 13; the prepared DESs generally have a decomposition temperature range from 150°C to 190°C. ILs that have the highest decomposition temperature are ChOH butyric acid and ChOH malic acid at 190°C followed by ChOH acetic acid and propionic acid with decomposition temperature of 175°C, and ChOH citric acid has the lowest decomposition temperature of 150 °C.

4.4 Viscosity Measurements for Choline Based ILs with Choline Hydroxide

The viscosity of ILs is a crucial factor influencing their performance as electrolytes for lithium ion batteries. Lower viscosity directly facilitates greater mobility of charge-carrying lithium ions within the electrolyte. This enhanced mobility leads to higher ionic conductivity. Conversely, highly viscous ILs impede lithium ion movement, leading to reduced ionic conductivity and consequently, poorer battery performance in terms of power output and efficiency. Additionally, high viscosity can hinder the uniform formation of the solid electrolyte interphase (SEI) layer on the anode, a critical barrier that protects the anode from degradation and promotes battery longevity. Therefore, optimizing the viscosity of IL-based electrolytes is essential for achieving desirable electrochemical properties and extended cycle life in lithium ion batteries. The viscosity measurements of the synthesized choline ILs with choline hydroxide were measured at 50 °C in this work; the results are in the Table 6. ChOH malic acid and ChoH citric acid show high viscosity compared to carboxylic based ILs. The viscosity values observed in Table 6 need to be contextualized within the range considered optimal for ionic liquids in gel polymer electrolytes (GPEs). For lithium-ion batteries, an ideal electrolyte should have low enough viscosity to allow for efficient ion transport while maintaining sufficient mechanical stability. Based on literature, a desirable viscosity range for GPEs is generally between 10-100 cP at operational temperatures (Karuppasamy *et al.*, 2017, 2020; Gupta, Jain and Tripathi, 2021). This range ensures adequate ionic conductivity without compromising electrolyte stability. If the viscosity is too high, it may hinder ion mobility, leading to reduced battery performance. On the other hand, too low a viscosity could result in leakage or structural failure. Therefore, the observed values of synthesized GPEs, while slightly higher than ideal, indicate that further optimization might be needed to reduce viscosity to the optimal range.

Table 6. Viscosity measurements of choline ILs based on choline hydroxide at 50ºC.

ILs	Viscosity (cP)
ChOH malic acid	1541
ChOH citric acid	18840
ChOH acetic acid	24.8
ChOH propionic acid	25.7
ChOH butyric acid	Not measured

4.5 Feasibility of Choline ILs Based on Choline Hydroxide for Use as Lithium ion Battery Electrolytes

Initial characterization of choline ILs synthesized from choline hydroxide and organic acids investigated viscosity, thermal stability, and state at room temperature.

Results indicate that these choline ILs may not be suitable as electrolytes. While FTIR confirms successful synthesis of the choline IL structure, their thermal stability (150-190 °C) is moderately satisfactory compared to other choline based ILs that have been utilized as electrolytes, which can withstand temperatures around more than 300°C (Yong *et al.*, 2016). Additionally, the high viscosity of ChOH malic acid and ChOH citric acid hinders potential large-scale application. ChOH with acetic acid and propionic acid solidified at room temperature, further disqualifying them as electrolytes. These results align with some literature findings; for example, the melting point of ChOH acetic acid varies between 51-72 °C across different studies (Fukaya *et al.*, 2007; Muhammad *et al.*, 2012; Cheng *et al.*, 2014), likely due to purity differences. In conclusion, choline based ILs require modifications to achieve higher thermal stability, lower viscosity, and a liquid state at room temperature for electrolyte applications.

4.6 Structural Characterization of Choline ILs from Choline TFSI and Organic acids

The synthesis of choline based ionic liquids with choline TFSI have undergoes two version of synthesis. Figure 14 illustrates the modified synthesis route (version 2) for choline based ionic liquids using choline TFSI, demonstrating improvements over the original method (version 1). Key modifications and their rationale include:

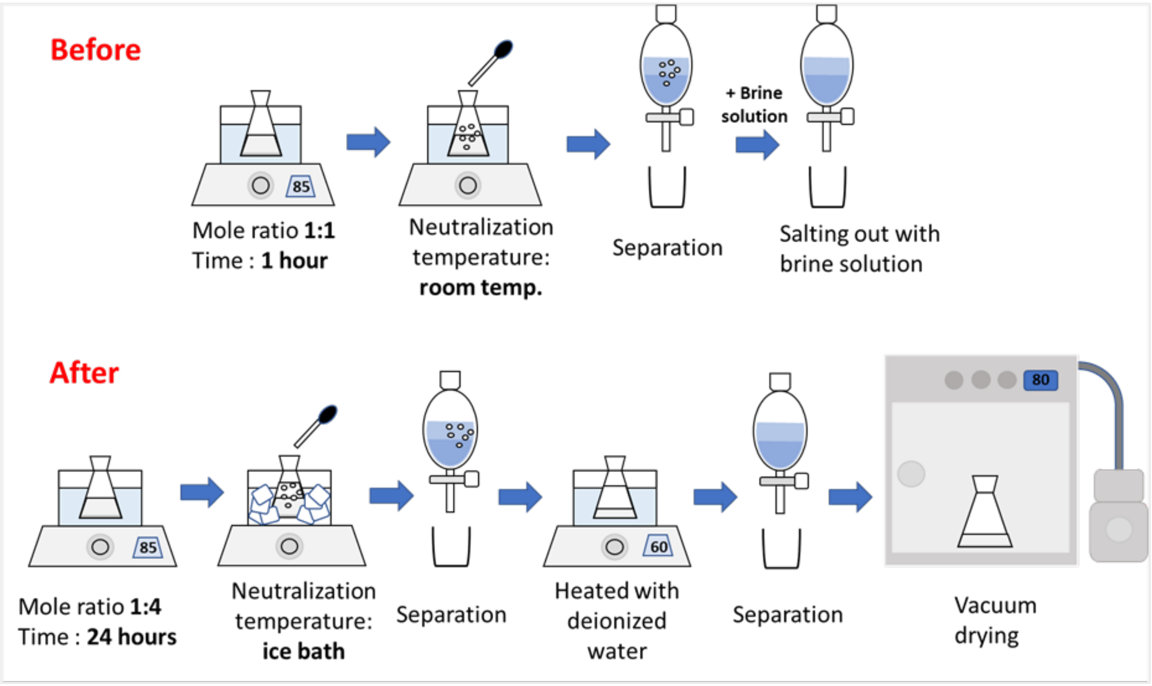


Figure 14. Schematic flow diagram of the synthesis of choline based ionic liquids with choline TFSI, before and after.

- Mole Ratio Adjustment (1:4): Increasing the carboxylic acid ratio drives the esterification equilibrium towards product formation, minimizing unreacted starting materials and potential impurities.

- Extended Reaction Time (24 hours): Ensures reaction completion, promoting higher product yields.

- Cooled Neutralization: Utilizing a cooled sodium carbonate solution mitigates excessive effervescence and further favors product formation according to Le Chatelier's principle (exothermic reaction).

- Purification with Deionized Water: Removes unreacted precursors, enhancing ionic liquid purity.

- Vacuum Drying (80 °C, 24 hours): Thoroughly removes residual water, guaranteeing a pure final product.

These optimizations in version 2 likely result in a more efficient and pure synthesis of choline-based ionic liquids. Supporting evidence for these improvements should be presented in subsequent sections using FTIR and NMR.

4.6.1 Nuclear Magnetic Resonance (NMR)

Proton and carbon nuclear magnetic resonance (NMR) spectroscopy are powerful techniques that can be used to determine the chemical structure of a molecule. Proton NMR provides information about the chemical environment of the protons in a molecule. The number of signals and their position in the spectrum give information about the number and types of protons present and their relative chemical environment (Damodaran, 2016). Protons in different environments, such as those attached to different functional groups or positions within the molecule, have different chemical shifts. The integration of each signal gives the relative number of protons in each environment, while the coupling patterns between neighbouring protons provide information about their connectivity. By comparing the NMR spectrum of an unknown compound to known spectra in a database or in the literature, it is possible to identify the compound and determine its structure.

In this study, carbon and proton NMR techniques were employed to verify the purity and structure of the synthesized ionic liquids. Figure 15A depicts the proton NMR spectrum of the initial synthesis of acetyl choline TFSI using the first methodology. This spectrum reveals the presence of both the intended structure (choline ester IL, indicated with blue) and unreacted starting material (choline TFSI), indicated by red alphabet labels.

However, Figure 15B, representing the product of synthesis method 2, demonstrates a significant improvement. The absence of the proton signal associated with the alcohol group in choline (at approximately δ = 5.3 ppm (Delso *et al.*, 2019)) indicates a more complete conversion of choline TFSI to the desired ester structure. Furthermore, carbon NMR analysis (Figure 15C) confirms successful synthesis with the characteristic ester group (-COO) peak at δ = 169.84 ppm (Simeral and Maciel, 1974). All proton and carbon signals are labelled with alphabets in Figure 15, including those specific to the $-N(CH_3)_3$ group in choline and the $-CF_3$ group in TFSI. The complete proton and carbon NMR spectra of all the synthesized choline ILs with choline TFSI will be included in the appendix at the end of this thesis (Figure S1).

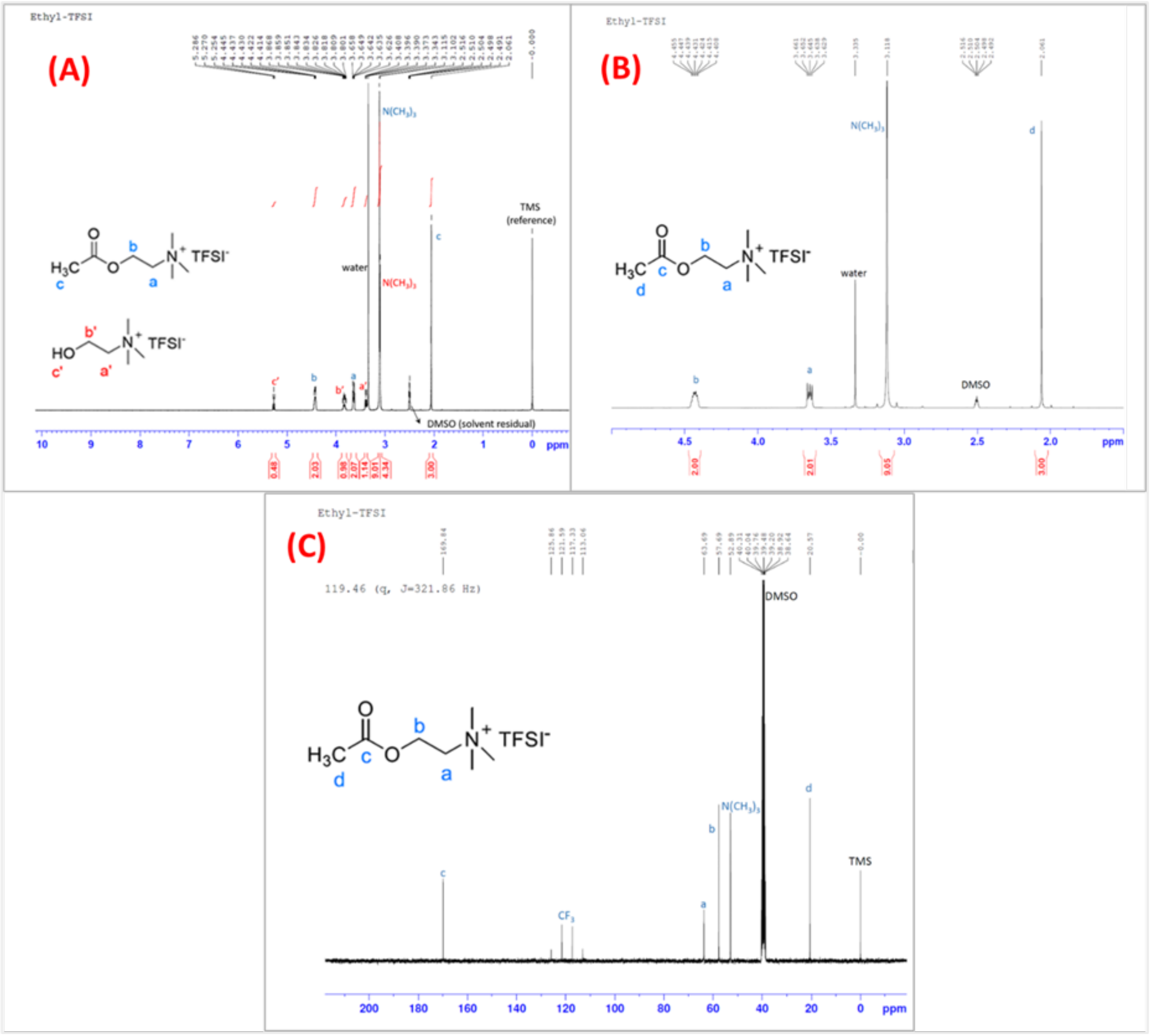


Figure 15. NMR spectrum of (A) C^{13} NMR spectrum of acetylcholine TFSI with synthesis method version 1, (B) C^{13} NMR and , (C) H^{1} NMR spectrum of acetylcholine TFSI spectra of acetylcholine TFSI with synthesis method version 2.

4.6.2 Fourier Transform Infrared (FTIR) Spectroscopy

The infrared (IR) spectrum provides valuable information about the functional group present in the synthesized compounds. In this study, Fourier transform infrared (FTIR) analysis was used to assess the success of synthesizing novel choline-based ionic liquids and to identify specific functional groups. Figure 16 shows that all the synthesized ILs (acetyl choline TFSI, propionyl choline TFSI, butyryl choline TFSI) and pristine choline TFSI, the synthesized ILs particularly exhibit absorption in the 1700-1760 cm^{-1} region, indicating the presence of a C=O stretch due to the ester group (-COO-) that was successfully introduced via esterification (Xie, Shao and Liu, 2010). Furthermore, the absorption at ~3600 cm^{-1}, corresponding to the hydrogen-bonded O-H stretch (Athokpam, Ramesh and McKenzie, 2017), was observed only in pristine choline TFSI, indicating that the alcohol functional group in choline was successfully converted into an ester group. Multiple weak peaks observed at ~3000 cm^{-1} were assigned to the stretching vibration of C-H (Moumene *et al.*, 2014), while the fingerprint region (<1500 cm^{-1}) (Nie *et al.*, 2021) exhibited no changes in the main structure of choline TFSI.

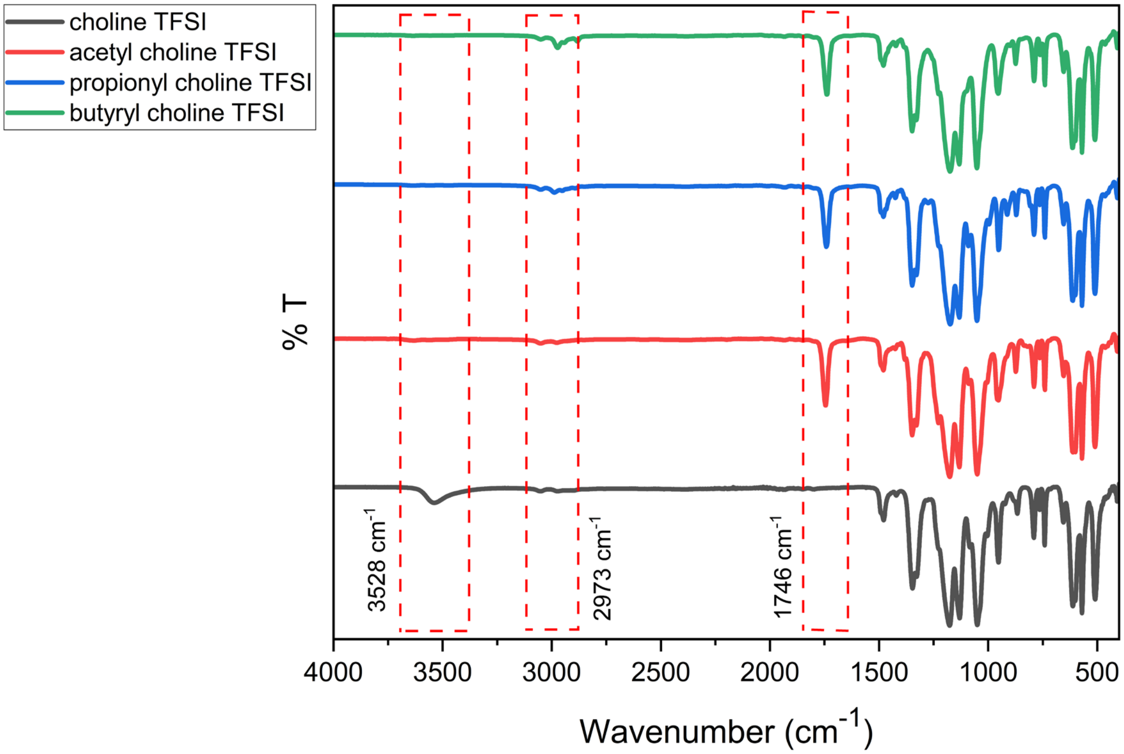


Figure 16. FTIR spectra of choline TFSI and synthesized choline based ionic liquids.

4.7 Thermal Analysis for Synthesized Choline Ester ILs and fabricated GPEs

4.7.1 Differential scanning calorimetry (DSC)

Differential Scanning Calorimetry (DSC) is a thermoanalytical technique that measures the heat flow into or out of a sample as it undergoes a controlled temperature change. This reveals various thermal transitions that a material experiences. In the realm of ionic liquid research, DSC is invaluable for determining properties like the glass transition temperature (Tg), where the ionic liquid shifts from an amorphous solid to a more fluid-like state. It can also detect melting points (Tm), crystallization events (Tc), and even decomposition temperatures. Understanding these thermal behaviors is crucial for assessing the stability, potential phase changes, and suitability of ionic liquids for applications where temperature plays a significant role. In this work, the synthesized ILs and pristine choline TFSI were analyzed using DSC to investigate their thermal transition profiles. The DSC thermogram (Figure 17) of pristine choline TFSI revealed three endothermic peaks: two at -7 °C and 2 °C, representing solid-to-solid transitions, and a broader peak around 22 °C (15-28 °C), indicating the melting point. These results align well with literature findings, which also report three peaks and melting points within the range of 25-33 °C (Villanueva *et al.*, 2015; Haddad *et al.*, 2024). This comparatively high melting point suggests that pristine choline TFSI may not be ideal as a lithium ion battery electrolyte. In contrast, the synthesized choline ester ILs exhibited no transformative peaks in their DSC thermograms. This absence of peaks is likely due to the large, asymmetric structure of choline ester ILs, which hinders the formation of stable crystalline structures.

Consequently, these ILs tend to exist in amorphous glassy states rather than crystallizing (Lobo Ferreira *et al.*, 2019).

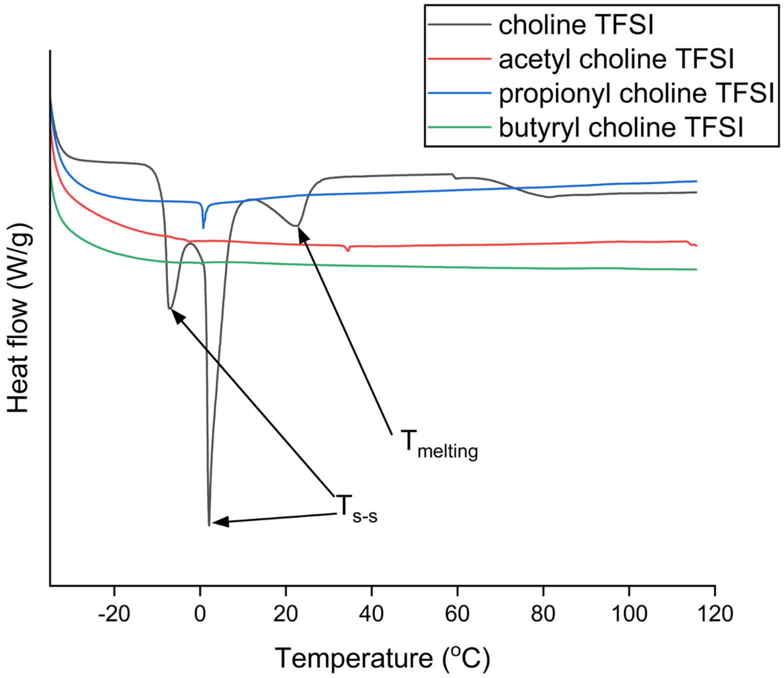


Figure 17. DSC thermogram of synthesized choline ester ILs and pristine choline TFSI

4.7.2 Thermogravimetric Analysis (TGA)

Thermal stability is a critical factor affecting the performance of gel polymer electrolytes in various energy storage devices. TGA is commonly used to assess the thermal stability of these electrolytes. Gel polymer electrolytes are engineered to balance high ionic conductivity, thermal stability, and electrochemical stability to improve the efficiency and safety of batteries and supercapacitors (Pandey *et al.*, 2016; Biria *et al.*, 2020; Pan, Wang and Fan, 2022). Studies have investigated various strategies to enhance the thermal stability of gel polymer electrolytes. For example, cellulose aerogel membranes as a framework for gel polymer electrolytes have been proposed to enhance the thermal stability of lithium ion batteries (Wan *et al.*, 2017). Moreover, the inclusion of specific additives like glycerin has been studied to alter the thermal characteristics of gel polymer electrolytes(Ali, Kareem and Polu, 2022). For this work, TGA was also employed to characterize the thermal stability of all the synthesized ionic liquids and fabricated ILGPEs. Figure 18A shows that the three synthesized choline ester ILs have a lower degradation temperature of 300 ºC compared to pristine choline TFSI, around 350 ºC. This might be due to increased organic content in the ILs or the side chain of cation and lead to a plasticization reaction, where the material's structure is altered and directly affects its thermal and mechanical properties and a reduction rigidity, deformation temperature, and thermal stability (Noorhisham *et al.*, 2021). More, all the fabricated GPEs are also subjected to a thermal stability test. Figure 18B shows all the GPEs were heated from room temperature to 500 ºC, and it was noticed that all the GPEs started to decompose around 300 ºC, similar to pristine ionic liquids. This decomposition temperature is also verified using derivative thermogravimetric analysis, where the rate of change of the sample mass with respect to temperature is measured. In Figure 18C, we notice the peak start at around 300 ºC where the GPEs start to decompose and reduce in weight significantly.

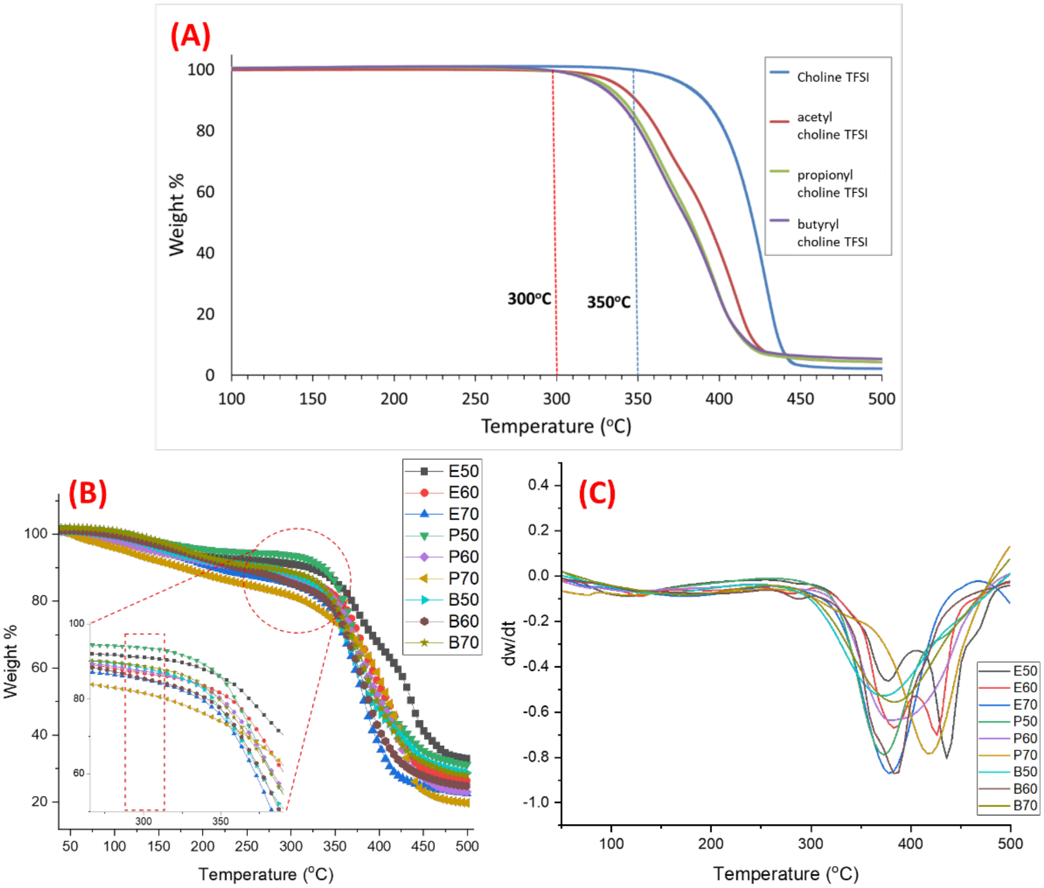


Figure 18. Thermogram of (A) choline TFSI and synthesized choline ionic liquids,(B) choline based ionic liquid GPE with 50%, 60%, and 70 % ionic liquids. (C) Derivative Thermogravimetry of choline based ionic liquid GPE with 50%, 60%, and 70 % ionic liquids.

4.8 Flammability test

Flammability testing of gel polymer electrolytes (GPEs) in lithium batteries is crucial for assessing safety risks and ensuring robust battery design. GPEs, despite offering advantages like enhanced ionic conductivity and flexibility, often contain flammable organic solvents(Ren *et al.*, 2021). These solvents pose a potential fire hazard, particularly in the event of battery damage or thermal runaway. In Figure 19, the snapshots show that the commercial battery separator ignited spontaneously and shrank (Figure 19D). Conversely, the fabricated GPE will not be ignited or inflammable. This is due to the low volatility of ionic liquids and their thermal stability towards heat. This shows the feasibility of utilizing choline-based ionic liquid as a safer alternative for lithium ion batteries electrolytes.

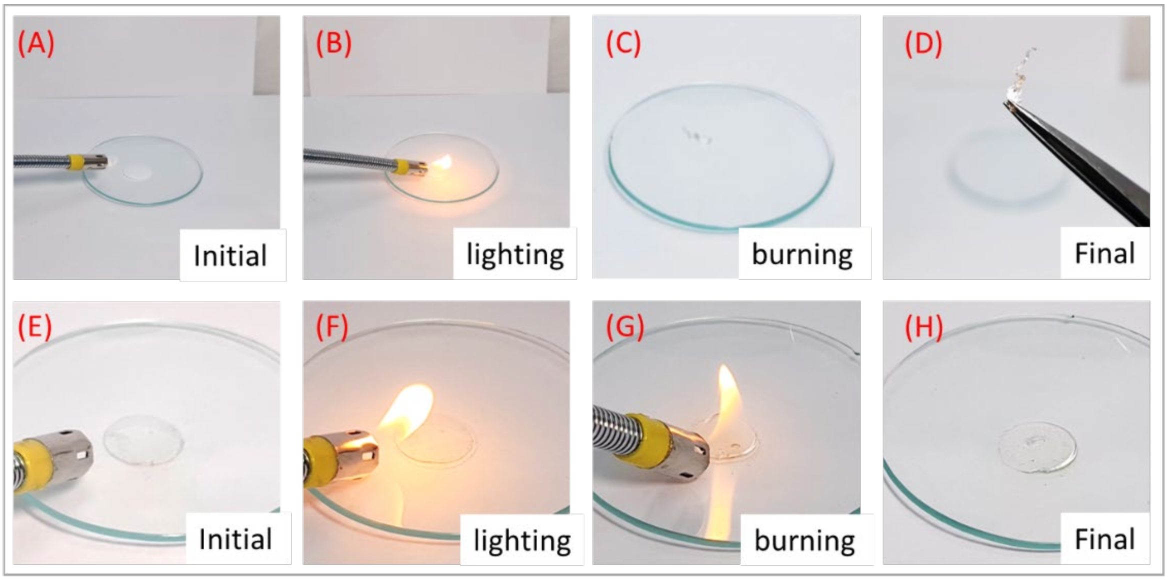


Figure 19. Snapshots of flammability test of a (A-D)commercial separator and (E-H) gel polymeric electrolyte with PVDF-HPF as the polymer.

4.9 Electrochemical Properties

4.9.1 Electrochemical Impedance Spectroscopy (EIS)

Ionic conductivities are essential to evaluate electrolyte performance for lithium battery usage, especially for GPE. Higher ionic conductivities indicate better ion mobility and conduction in electrical current. Different ratios of ionic liquid to PVDF-HFP were investigated to improve ionic conductivity while seeking a balance with the polymer's advantageous properties. Ionic conductivities were obtained by combining the Nyquist plot results (Figure 20A) and equation (2). The findings presented in Table 7 demonstrate the relationship between the ionic conductivity of the fabricated Gel Polymer Electrolytes (GPEs) and the varying ratios of Ionic Liquids (ILs) to polymer. Overall, an increase in the proportion of ionic liquid within the GPEs corresponded to higher ionic conductivities. Notably, this trend was consistently observed for butyryl choline TFSI and propionyl choline TFSI. However, for acetyl choline TFSI, a slight decrease in conductivity was noted when the IL composition increased from 60 wt% to 70 wt%. The GPE comprising 70 wt% propionyl choline TFSI is of particular significance, which exhibited the highest ionic conductivity recorded at 6.09 x 10^{-2} Scm^{-1}. Conversely, acetyl choline TFSI consistently exhibited the lowest ionic conductivities across all compositions evaluated. Ionic conductivity and Electrochemical Impedance Spectroscopy (EIS) measurement of ionic liquids are intricately influenced by the molecular structure, particularly the alkyl chain length and functional groups in their constituent ions (Nordness and Brennecke, 2020). Longer alkyl chains within the ionic liquid molecules generally lead to increased viscosity (Gaciño *et al.*, 2011). This higher viscosity impedes the movement of ions within the liquid, thus reducing the overall ionic conductivity (Yuan *et al.*, 2018). The elongated alkyl chains introduce more steric hindrance and stronger intermolecular interactions, which hinder the mobility of ions, thereby decreasing conductivity (Liu, Zhang and Zhang, 2018). Conversely, shorter alkyl chains or smaller substituents tend to lower viscosity. This reduction in viscosity facilitates greater freedom of movement for ions within the liquid, resulting in higher ionic conductivities (Ramesh and Ling, 2010). Shorter chains experience less steric hindrance and weaker intermolecular interactions, allowing ions to move more freely and efficiently (Y. L. Wang *et al.*, 2020). Additionally, the type and distribution of functional groups in the ionic liquid molecules play a crucial role in determining their conductivity properties as well. Polar functional groups, such as hydroxyl (-OH) or amino (-NH_2) groups, can enhance the solvation of ions and thus increase the conductivity by facilitating ion transport (Li *et al.*, 2024).

On the other hand, the ratio of ionic liquids to polymers in gel polymer electrolytes significantly influences the ionic conductivities of the electrolytes. Optimal composition can lead to improved ionic conductivities, enhancing the overall performance of the electrolyte system (Sun *et al.*, 2016; Iacob *et al.*, 2017; Jeon *et al.*, 2020). Studies have demonstrated that the molar ratio of ionic liquids to polymers can significantly impact the ionic conductivity of gel polymer electrolytes. By adjusting this ratio, researchers have achieved higher ionic conductivities, which are crucial for efficient energy storage device operation (Shin *et al.*, 2016; Xiaoen Wang, Girard, *et al.*, 2019; Z. Liu *et al.*, 2021). The selection of an appropriate composition is vital for balancing interactions between ionic liquids and polymers and optimizing the ionic transport properties within the electrolyte system (Gupta *et al.*, 2021; Yamada, 2022; Polu *et al.*, 2023).

The choice of polymer in an ionic liquid gel polymer electrolyte (ILGPE) also significantly influences how composition affects ionic conductivity. Polymers like PVDF-HFP, with polar groups, interact favorably with ionic liquids (Ruan *et al.*, 2022). Increasing ionic liquid content in these systems generally boosts conductivity due to better ion dissociation and increased polymer flexibility (Ye, Rick and Hwang, 2013). In contrast, polymers like PMMA have weaker interactions with ILs (T. Gao *et al.*, 2021), so increasing IL content may not yield the same conductivity improvements and could even lead to phase separation. Similarly, the type of ionic liquid also plays a critical role. ILs with coordinating anions (like TFSI$^-$ or FSI$^-$) promote lithium ion dissociation, increasing the number of free charge carriers and enhancing conductivity (K. Liu *et al.*, 2021b).

Table 7. Ionic conductivities of choline based ionic liquid GPE with different compositions.

GPE composition (wt%)(C1:C2)			**Ionic conductivities σ ($\times10^{-2}$ Scm^{-1})**
C1 (ILs)		**C2 (PVDF-HFP)**	
acetyl choline TFSI	50	50	0.59
	60	40	0.79
	70	30	0.71
propionyl choline TFSI	50	50	1.24
	60	40	1.86
	70	30	6.09
butyryl choline TFSI	50	50	1.38
	60	40	1.99
	70	30	3.93

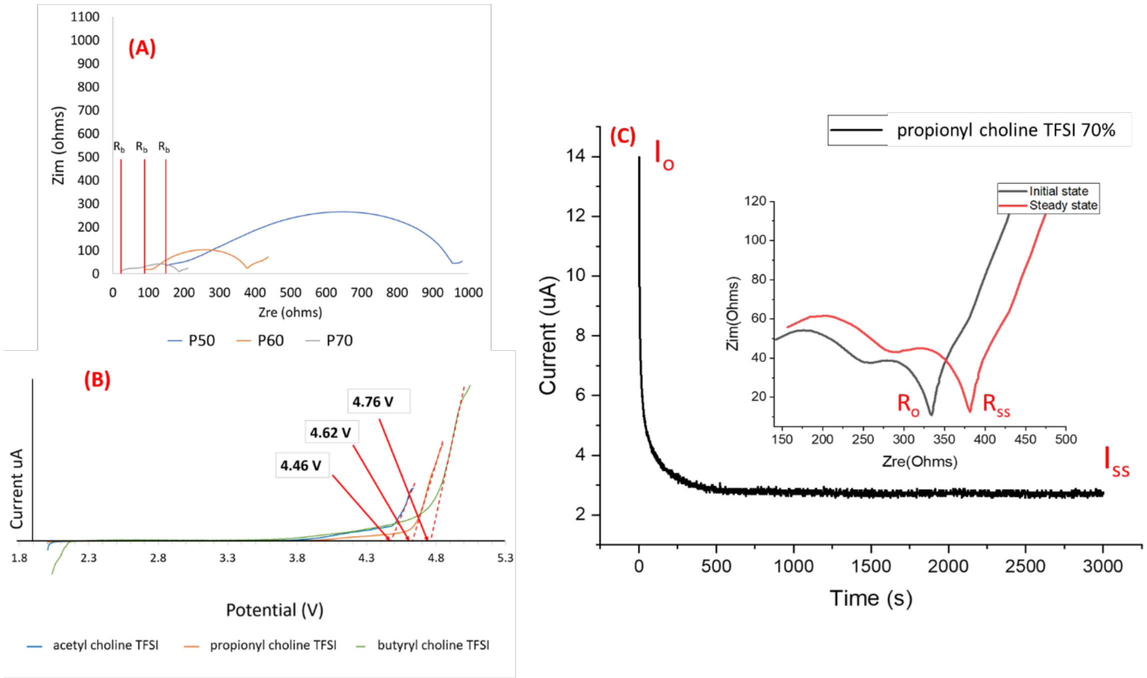


Figure 20. (A) Nyquist plot of propionyl choline TFSI GPEs (50,60,70 %), (B) Linear sweep voltammogram of synthesized choline based ionic liquids (C) DC Polarisation curve obtained via chronoamperometry for propionyl choline TFSI 70 samples. (Inset Nyquist curve of same sample before and after dc polarisation).

4.9.2 Linear Sweep Voltammetry (LSV)

The electrochemical stability window is another aspect of evaluating an electrolyte's performance in a lithium battery. An electrochemical stability window (also known as the electrochemical window) is the range of potentials or voltages in which an electrolyte or an electrochemical system can remain stable and not undergo any chemical or electrochemical reactions, potentially compromising battery safety through the formation of reactive byproducts and increasing the risk of thermal runaway (Snyder and Theis, 2022). Furthermore, degradation reactions outside the ESW can damage electrode-electrolyte interfaces, hindering lithium ion transport and leading to capacity fade and reduced cycle life (A. Wang *et al.*, 2018). In Figure 20B, the synthesized ionic liquids start to decompose above 4.46 V (acetyl choline TFSI), 4.62 V for propionyl choline TFSI, and butyryl choline TFSI has the highest electrochemical stability window (4.76 V) among the three ionic liquids. However, the three synthesized ionic liquids are suitable for utilization as a lithium ion battery electrolyte since the common operating voltage for a typical lithium ion battery ranges from 3-4.3 V(Saïdi *et al.*, 2003; You *et al.*, 2020). The synthesized ionic liquids in this study demonstrate promising ESWs, with butyryl choline TFSI exhibiting the widest stability window of 4.76 V. The wider ESWs observed, particularly for butyryl choline TFSI, suggest these electrolytes could enable the use of higher-voltage cathode materials. This flexibility in cell design could potentially increase the overall energy density of the batter (Yang *et al.*, 2023). However, it's important to note that while a wide ESW is essential, it does not guarantee long-term stability under realistic battery operating conditions. Further investigations into cycling stability and the potential influence of electrode materials on electrolyte degradation are necessary to assess the practical viability of these synthesized ionic liquids fully. Additionally, studies exploring the temperature dependence of the ESW would be valuable, as extreme temperatures can impact electrolyte stability (D. Luo *et al.*, 2021). The potential use of additives or surface modification strategies to further enhance the ESW of these ionic liquids also warrants investigation, particularly if the goal is to push the limits of high-voltage battery design (Rüther *et al.*, 2020).

4.9.3 Transference number

Transference number is a measurement of the relative mobility of ions in an electrolyte. In the context of lithium ion batteries, the transference number refers to the fraction of the total current carried by lithium ions in the electrolyte during the battery's charging and discharging cycles (Buss *et al.*, 2017). The transference number is an important characteristic of a lithium ion battery's electrolyte because it affects its performance and safety. A low transference number can lead to uneven current distribution within the battery, which can cause side reactions and reduce the battery's efficiency(Zhao *et al.*, 2021). A low transference number can also increase the risk of dendrite formation, which can short-circuit the battery and cause it to catch fire or explode (Q. Liu *et al.*, 2022). In this work, the GPE is subjected to DC polarisation and the change in the bulk resistance in the electrolyte is also measured using AC electrochemical impedance spectroscopy method (Figure 20C). Table 8 reveals that propionyl choline TFSI (70%) exhibits the highest ionic conductivity among the synthesized GPEs and possesses a high transference number of 0.88. Conversely, butyryl choline TFSI (60%) demonstrates the highest transference number (0.94), while acetyl choline TFSI (70%) has the lowest (0.85). Notably, all synthesized GPEs exhibit transference numbers significantly exceeding those typically reported in the literature (0.2-0.5)(Park *et al.*, 2013; Zhao *et al.*, 2021). Transference numbers above 0.55 are generally considered high (Cheng *et al.*, 2018; Halder, Mohamed, S. W. Kuo, *et al.*, 2024)**.**

While the transference numbers obtained for the synthesized GPEs appear to be significantly higher than those typically reported in the literature, it is important to note a potential source of error in the measurements. Specifically, the initial current measurement was not conducted in the appropriate timeframe; the first measurement should have been captured before 0.01 seconds. This delay in capturing the initial current can affect the accuracy of the transference number calculation, as it influences the determination of the initial current (Io) and steady-state current (Iss), both of which are critical for the calculation. As a result, the reported transference numbers, while promising, should be regarded with caution. To ensure the validity of these results, further experiments should be conducted with corrected timing to verify the high transference numbers observed in this study.

Table 8. Transference number of fabricated ionic liquid GPEs and its relative value for transference calculation.

GPEs	I_{ss} (x10^{-5} A)	I_o(x10^{-5} A)	mV	R_o(ohm)	R_{ss}(ohm)	t_{li+}
Acetyl choline TFSI 70%	0.2616	1.028	10	283.9	335.6	0.85
Propionyl choline TFSI 70%	2.75	13.98	10	334	381	0.88
Butyryl choline TFSI 60%	0.487	2.061	10	359	380	0.94

4.9.4 Cyclic voltammetry (CV)

Cyclic voltammetry (CV) is an electrochemical technique used to study the redox behaviour of materials in solution. In CV, a potential is applied to a working electrode while the current flowing through the circuit is measured. The potential is swept over a range of values, typically from a negative value to a positive value and then back again. This creates a cyclic voltammogram, a plot of the current versus the applied potential. This technique also gives helpful information on the type of electrochemical system, allowing the researcher to determine what type of charge is being stored. Figure 21 (A-C) shows the CV profile of the electrochemical double layer, pseudo capacitor, and battery. The electric double layers will show rectangular CVs, while electrochemical systems with intense and distinct oxidation and reduction peaks are categorized as faradaic or battery (Schoetz *et al.*, 2022). Pseudo capacitor, however, has a feature of a combination of the two electrochemical systems. For this work (Figure 21D), the GPE of propionyl choline TFSI 70 was sandwiched between an LCO electrode and Li metal and cycled between 3 V and 4.5 V with a scan rate of 0.1 mVs^{-1}. The consistent oxidation and reduction peak potentials observed over five cycles demonstrate the material's stability during repeated lithiation (insertion of lithium ions) and delithiation (extraction of lithium ions) to and from the electrode, indicating its suitability for rechargeable lithium ion battery applications. Also, distinct oxidation and reduction peaks can be observed in the CV profile, indicating that the system behaves like a battery rather than a capacitor or pseudo-capacitor. While the CV results in Figure 21D suggest battery-like behavior rather than pseudocapacitive behavior, further analysis is required to clarify this. Specifically, information on the LCO particle size and a more detailed b-factor analysis using various scan rates for cyclic voltammetry (CV) is needed to better understand the charge storage mechanism. These additional tests would help confirm whether the system exhibits purely faradaic behavior or if there is a pseudocapacitive contribution. Therefore, additional experiments must be conducted to provide a more comprehensive evaluation of the electrochemical behavior.

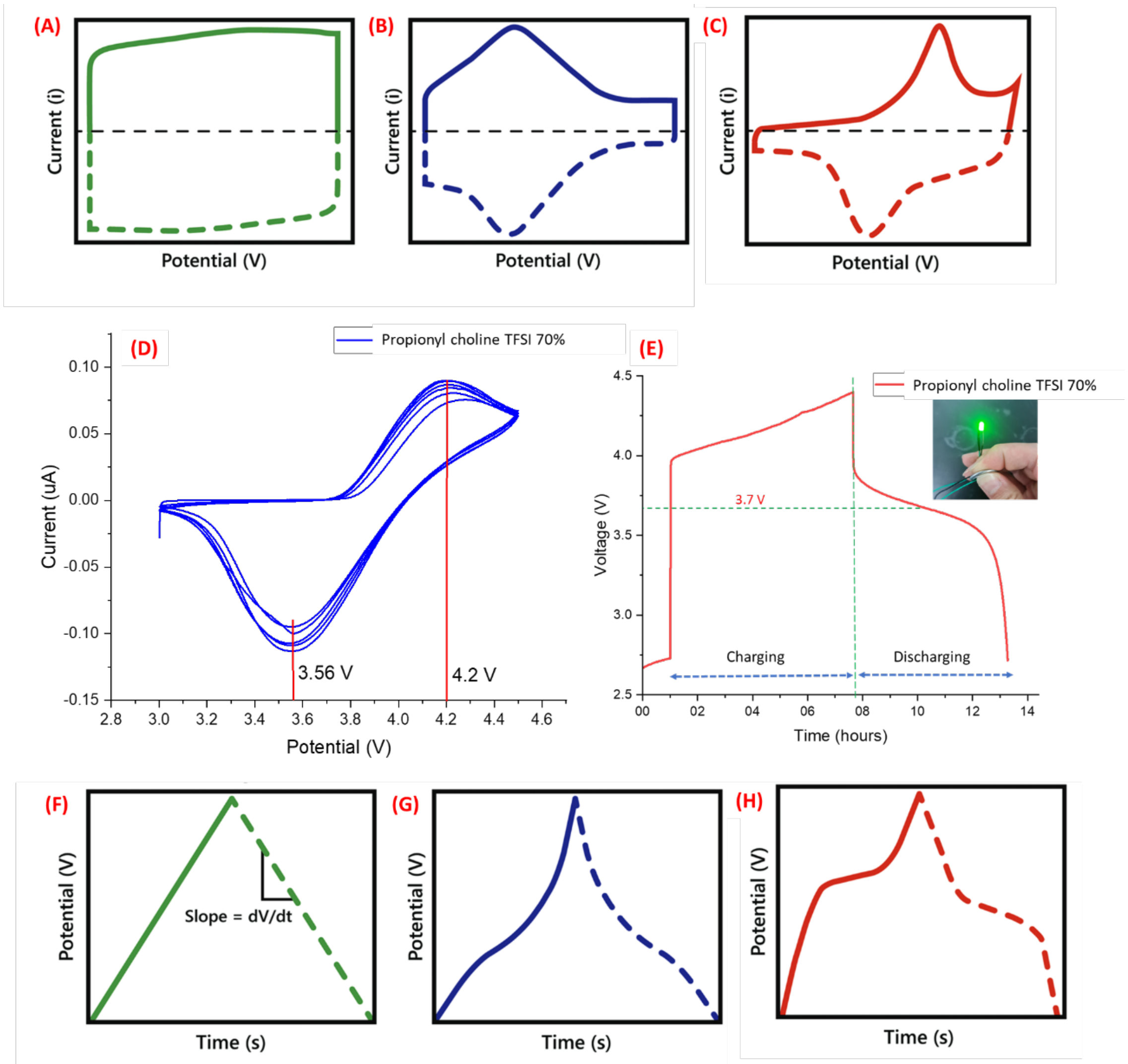

(A)
Current (i)
Potential (V)
(B)
Current (i)
Potential (V)
(C)
Current (i)
Potential (V)
(D)
Propionyl choline TFSI 70%
Current (uA)
0.10
0.05
0.00
-0.05
-0.10
-0.15
3.56 V
4.2 V
2.8
3.0
3.2
3.4
3.6
3.8
4.0
4.2
4.4
4.6
Potential (V)
(E)
Propionyl choline TFSI 70%
Voltage (V)
4.5
4.0
3.7 V
3.5
3.0
2.5
Charging
Discharging
00
02
04
06
08
10
12
14
Time (hours)
(F)
Potential (V)
Slope = dV/dt
Time (s)
(G)
Potential (V)
Time (s)
(H)
Potential (V)
Time (s)

Figure 21. CV profiles of typical (A) electrical double layer (B) pseudocapacitive (C) battery (Mathis *et al.*, 2019) (D) GPE propionyl choline TFSI 70%. Galvanostatic charge-discharge curve of (E) Propionyl choline TFSI 70 GPE (Current density: 0.25mAcm^{-2}, Gravimetric capacity: 0.35 mAhg^{-1}) (inset: Green LED light up by coin cell assembled with fabricated GPE.), (F) electrical double layer (G) pseudocapacitive (H) battery(Mathis *et al.*, 2019).

4.9.5 Galvanostatic charge-discharge (GCD)

Galvanostatic charge-discharge is a method used in electrochemistry to characterize the behaviour of an electrochemical cell or battery. In this method, a constant current is applied to the cell or battery to either charge it (increase its stored energy) or discharge it (use its stored energy). Also, the shape of the GCD curve can be used to differentiate the electrochemical system from the electrical double layer, pseudocapacitive, and battery. An electrical double layer will have a linear relationship between charge and voltage, which results from the physical electrostatic attraction of charges. Thus, it exhibits linear charging responses to applied potentials (Mathis *et al.*, 2019), as shown in Figure 21F. GCD curves for batteries typically show plateaus of constant potential associated with the redox reactions in the peak potential in the battery-type system (Figure 21H). In this work, the GCD shows prominent plateaus in the charging and discharging curve, as seen in Figure 21E. In the GCD curve, the assembled battery has a nominal voltage of 3.7 V, a typical nominal voltage for LCO batteries (Kim *et al.*, 2009; Krieger, Cannarella and Arnold, 2013; Stan *et al.*, 2014). Nominal voltage can be described as a typical operating zone, rather than a fixed value. The actual voltage of an LCO battery depends on how much charge it holds. A fully charged LCO battery will be closer to 4.2 volts (Wu *et al.*, 2019), and as it discharges for use, the voltage will gradually decrease. To prevent damage, the battery shouldn't be discharged below a certain voltage, typically around 3.0 volts (Wan *et al.*, 2021). There can also be slight variations in the nominal voltage depending on the specific makeup of the LCO and any additives used in its construction (L. Wang *et al.*, 2018). Additionally, factors like how fast the battery discharges (C-rate) and the surrounding temperature can also slightly influence the voltage you see (Spitthoff, Shearing and Burheim, 2021). The inset in Figure 21E shows a green LED lit by the fabricated coin cell battery.

4.10 Mechanical properties

Mechanical properties play a vital role in the safety and performance of lithium ion batteries. Robust mechanical strength in electrolytes, particularly gel polymer electrolytes, is crucial for suppressing lithium dendrite growth. Dendrites, which form on the anode surface during battery cycling, can pierce the electrolyte and lead to internal short circuits, compromising safety and severely degrading battery performance (Aslam *et al.*, 2021). A mechanically robust GPE can physically resist dendrite penetration and maintain the integrity of the separator layer (Y. Li *et al.*, 2021). Figure 22 shows the fabricated GPE in this work. The GPE was stretched with tension force, and the GPE was elongated and doubled its original length from 3 cm to 6 cm without tearing, which shows the GPE has good tensile strain. This result aligns well with reported values for other GPEs, which typically exhibit elongation at break percentages around 200%. (Sun *et al.*, 2000; Gu, Liu and Liu, 2020; Tamainato *et al.*, 2022).

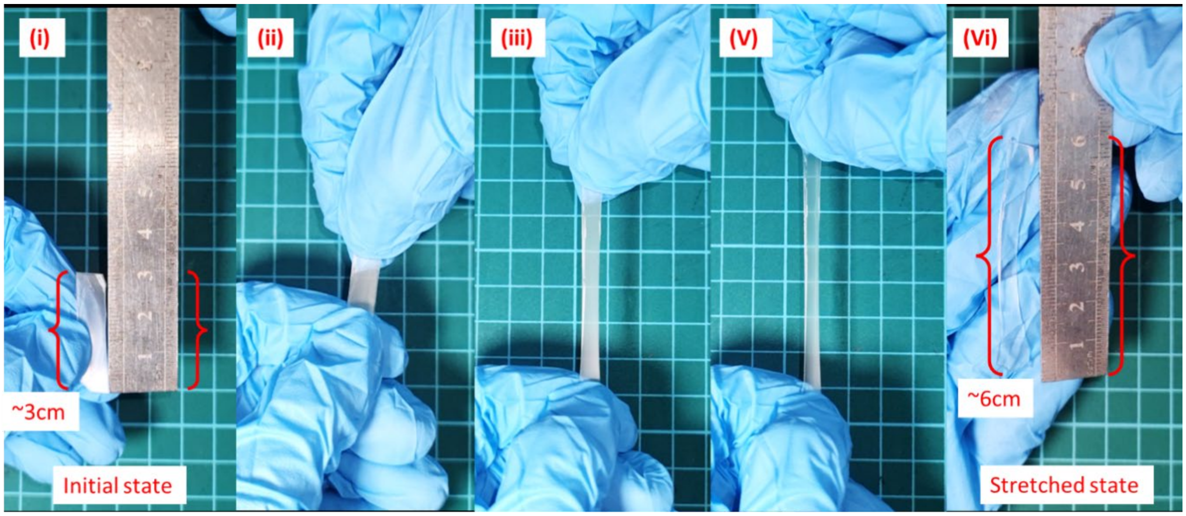


Figure 22. Photographs of fabricated ionic liquid GPE in the (i) initial state and after (ii) stretched state.

4.11 The Significant of Cation and its Alkyl chain length

In this study, the cations were synthesized using organic acids—acetic acid, propionic acid, and butyric acid—leading to an increase in alkyl chain length from C2 (acetic acid) to C4 (butyric acid). This variation in alkyl chain length has a significant impact on the physical and electrochemical properties of the resulting choline-based ionic liquids.

- Effect on Ionic Conductivity: As the alkyl chain length increases, the overall viscosity of the ionic liquid tends to increase due to the larger, bulkier structure of the cation. A longer alkyl chain introduces more van der Waals interactions, which can hinder the free movement of ions and reduce ionic conductivity (Hu and Peng, 2014). This is evident in this study, where acetyl choline TFSI (with the shortest chain, C2) generally exhibited the lowest viscosity and highest ionic conductivity, while butyryl choline TFSI (C4) had the highest viscosity and lower ionic conductivity. This relationship is well-supported in the literature, where shorter-chain ILs often outperform longer-chain counterparts in terms of conductivity.

- Impact on Electrochemical Stability: The alkyl chain length also influences the electrochemical stability of the ionic liquid. Shorter alkyl chains, like those in acetyl choline TFSI, provide greater stability in the electrochemical window due to reduced steric hindrance and better solvation of ions (Fan *et al.*, 2022). However, longer alkyl chains can lead to improved hydrophobicity, which can stabilize the ionic liquid at higher voltages but may reduce overall electrochemical performance due to slower ion mobility (Montanino *et al.*, 2011).
- Influence on Gel Polymeric Electrolytes (GPEs): When these choline-based ILs are incorporated into the GPEs, the varying alkyl chain lengths affect both the mechanical properties and ionic transport within the polymer matrix. Longer alkyl chains, like in butyryl choline TFSI, introduce more hydrophobic regions within the GPE, potentially improving the mechanical strength but at the cost of reduced ionic mobility (Markiewicz *et al.*, 2021). Conversely, acetyl choline TFSI, with its shorter chain, provides more efficient ionic transport but may result in a slightly less robust mechanical structure.
- Alkyl Chain Length and Lithium-ion Transference Number: The alkyl chain length can also affect the lithium-ion transference number. In this study, butyryl choline TFSI (C4) exhibited the highest transference number (0.94), suggesting that the longer alkyl chain improves the selective transport of lithium ions compared to shorter chains like acetyl choline TFSI (0.85). This is possibly due to the reduced anion mobility in longer-chain ILs, which allows a higher proportion of the total current to be carried by lithium ions (Moosavi *et al.*, 2016).

4.12 Summary

This study investigated the suitability of choline-based ionic liquids (ILs) as electrolytes for lithium ion batteries. Initially, choline hydroxide was used to synthesize ILs via neutralization with organic acids. However, these initial ILs exhibited properties (room temperature state, viscosity, and thermal stability) that limited their potential as electrolytes.

An alternative approach employed choline TFSI as the starting material and reacted it with ethanoic, propanoic, and butanoic acids through an esterification reaction. This method yielded refined products verified by FTIR and NMR spectroscopy. Subsequently, the electrochemical properties of these new ILs were evaluated, including ionic conductivity, electrochemical stability, redox capability, and lithium ion transference number.

The results revealed that a combination of propionyl choline TFSI and a polymer host, PVDF-HFP, at a 70:30 ratio demonstrated the most promising potential as a lithium ion battery electrolyte. Additionally, safety aspects like thermal and mechanical properties were assessed. The ionic liquids exhibited good thermal stability, degrading above 300 °C, and satisfactory tensile properties, potentially mitigating dendrite growth and short circuit risks.

Table 9 summarizes references utilizing ionic liquids and polymer matrices for lithium ion electrolytes. It highlights their key chemical properties and thermal stability for comparison with the results from this work. Notably, the gel polymer electrolyte (GPE) developed in this study achieves a high ionic conductivity and transference number while maintaining good thermal and electrochemical stability. These combined properties position it as a promising all-rounder for safe lithium ion battery electrolyte applications.

Table 9.Comparison of electrochemical properties and thermal properties between the literature's value and this work

Ionic liquid used in GPE	Ionic conductivities (mScm^{-1})	Electrochemical stability window (V)	Transference number	Decomposition temperature (^oC)	Ref.
B4MePyTFSI 1-butyl-4-methylpyridinium bis(trifluoromethanesulfonyl)imide	0.20	5.50	0.70	-	(Yang *et al.*, 2014)
FSI-TGPE bis(fluorosulfonyl)imide	0.22	4.60	-	221	(Martinez-Ibañez *et al.*, 2023)
PDEIm-*40%* poly(1,2-diethoxyethylimidazolium) bis(trifluoromethanesulfonyl)imide	1.78	4.20	0.42	400	(Hu *et al.*, 2020)
EMIMFSI 1-ethyl-3-methylimidazolium bis(fluoromethanesulfonyl)imide	0.38	4.70	0.40	200	(S. K. Singh *et al.*, 2018)
[BMIM]BF$_4$ 1-Butyl-3-methylimidazolium tetrafluoroborate	1.40	4.50	-	240	(Zhai *et al.*, 2014)
Pyr14TFSI 1-butyl-1-methylpyrrolidinium bis(trifluoromethanesulfonyl)imide	0.72	4.70	0.47	320	(Yuan *et al.*, 2023)
PYR$_{13}$FSI N-propyl-N-methylpyrrolidinium bis(fluoromethanesulfonyl)imide	1.60	4.30	0.43	240	(Singh, Dutta and Singh, 2020)
EMITFSI 1-Ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide	3.20	5.70	0.62	-	(M. Li *et al.*, 2018)
BMImNfO 1-butyl-3-methylimidazolium nonafluorobutanesulfonate	26.10 (100^{o}C)	4.90	0.44	336	(Karuppasamy *et al.*, 2016)

Acetyl choline TFSI 60 % 2-(Acetyloxy)-N,N,N-trimethylethanaminium bis(trifluoromethylsulfonyl)imide	7.88	4.46	0.85	300	This work
Propionyl choline TFSI 70% 2-(Propionyloxy)-N,N,N-trimethylethanaminium bis(trifluoromethanesulfonyl)imide	60.92	4.62	0.88	300	This work
Butyryl choline TFSI 70% 2-(Butanoyloxy)-N,N,N-trimethylethanaminium bis(trifluoromethylsulfonyl)imide	39.26	4.76	0.94	300	This work

Chapter 5: Conclusion, Future Work, and Their Potential Beyond

5.1 Conclusion

The ever-growing demand for safe and sustainable energy storage solutions has fuelled intensive research into advanced battery technologies. Safety concerns associated with traditional lithium ion batteries highlight the critical need for the development of novel electrolyte materials. This thesis addressed this challenge by focusing on the synthesis and incorporating choline-based ionic liquids (ILs) into polymer matric of PVDF-HFP as GPEs as promising candidates for safer and more efficient electrolytes. Successfully achieving all research objectives, a series of novel choline-ester ILs were synthesized using facile neutralization and esterification methods. Characterization techniques like NMR and FTIR confirmed their structures and purity, aiding in refining the synthesis process.

These ILs demonstrated favorable thermophysical and electrochemical properties tailored for lithium battery applications. Their high thermal stability (up to 300°C) and non-flammability enhance their potential as safe electrolytes. Additionally, the choline-ester ILs exhibited exceptional electrochemical properties: high ionic conductivities (up to 6.09 x10^{-2} Scm^{-2}), high oxidative stability (above 4.4 V, up to 4.76 V), and high transference numbers (above 0.85, up to 0.94). GCD results confirmed battery behaviour in a full LCO/graphite coin cell with a nominal voltage of 3.7 V. Preliminary mechanical tests indicated promising flexibility, with elongation up to 200% without failure.

The incorporation of these ILs into GPEs using a PVDF-HFP matrix significantly improved safety while maintaining electrochemical performance comparable to other ILGPE system. The structure-property relationships uncovered, particularly the effect of alkyl chain length on ionic conductivity, offer valuable guidance for further optimization. Future studies investigating a broader range of choline derivatives and advanced GPE architectures could unlock the full potential of these materials, enabling a new generation of safe, high-performance lithium-ion batteries and potentially extending their applications to other electrochemical energy storage devices.

5.2 Future Work: Building upon the Success of Choline-based GPEs

The promising results obtained in this thesis lay the groundwork for a rich array of future research endeavours to elevate the performance and scalability of choline based GPEs for lithium battery applications. Here's a detailed exploration of potential directions:

1. Electrochemical Performance Optimization

- Cycling Stability and Capacity Retention: In-depth cycling tests over hundreds or even thousands of cycles are crucial to evaluate long-term GPE stability within a full battery cell. Assessing capacity retention will reveal how well the choline based ILs mitigate capacity fading mechanisms.
- Electrode Compatibility and SEI Formation: Investigating the compatibility of the GPEs with different anode and cathode materials (graphite, silicon, high-nickel cathodes, etc.) is essential. Techniques like X-ray photoelectron spectroscopy (XPS) and scanning electron microscopy (SEM) can elucidate interfacial reactions and the nature of the solid electrolyte interphase (SEI) formed, which impacts cell lifetime.
- Lithium Salt Synergy: Exploring different lithium salts ($LiPF_6$, LiTFSI, LiFSI, and others) and their concentrations will uncover potential synergies with the choline based ILs. Understanding lithium salt dissociation behaviour within the GPE matrix, as it affects ionic conductivity and interfacial properties, is vital. Computational modelling (e.g., Molecular Dynamics simulations) can provide valuable insights into these interactions at the molecular level.

2. Advanced GPE Architectures

- Novel Polymer Matrices: While PVDF-HFP is a well-established GPE polymer, researching alternative bio-based polymers, functionalized polymers, and block copolymers could unlock even better mechanical properties, interfacial compatibility, and tailored ion transport mechanisms.
- Composite Polymer Electrolytes: Incorporating ceramic fillers (like SiO_2, Al_2O_3) or solid-state electrolyte particles could further enhance lithium ion conductivity, suppress dendrite growth, and improve mechanical robustness.
- In-Situ Polymerization: Directly polymerizing the GPE within the battery cell offers a promising route for seamless interfacial contact, potentially minimizing interfacial resistance and boosting performance.
- 3D Engineered Structures: Employing techniques like 3D printing or electrospinning could enable the fabrication of GPEs with controlled microstructures and tailored interfaces, potentially leading to optimized ion transport pathways and improved electrochemical stability.

3. Process Scalability and Refinement

- Optimized Synthesis of Choline-based ILs: Investigating alternative synthesis pathways, such as continuous flow methods or solvent-free reactions, could enhance the scalability of IL production. Strategies to reduce waste, lower energy consumption, and streamline purification will be crucial for industrial translation.
- Co-solvent Strategies: Exploring the use of carefully selected co-solvents can improve the GPE fabrication process. Co-solvents might aid in polymer dissolution, facilitate IL incorporation, and control GPE morphology, ultimately boosting performance.

- Roll-to-Roll Manufacturing: Adapting GPE fabrication techniques to roll-to-roll processes promises large-scale manufacturing capabilities, making the technology commercially viable.

4. Broader Explorations

- Temperature Dependence: Thoroughly analyzing the GPE's ionic conductivity and stability across a wide temperature range is essential for real-world battery applications under varying climates.
- Dendrite Suppression: The potential of choline based ILs and designed GPE microstructures to suppress lithium dendrite growth should be rigorously investigated. Combining experimental observations (SEM) with theoretical modeling can provide a comprehensive understanding of dendrite suppression mechanisms.
- Beyond Lithium ion: While the focus here was on lithium ion batteries, these choline-based GPEs could find potential applications in other electrochemical energy storage systems, such as sodium-ion, potassium-ion, or multivalent-ion batteries.

This extensive roadmap for future exploration positions choline based GPEs at the forefront of safe and high-performance electrolyte development. By systematically addressing these areas, this research has the potential to make a transformative impact on the future of energy storage.

5.3 Application Beyond Energy Storage

The exceptional properties of choline-based ionic liquids, including their biodegradability, adaptability, and non-toxicity, open a wide range of applications beyond their established use as electrolytes. Here are some key areas where their unique characteristics hold significant promise:

- Biomass Processing and Biorefineries: Choline-based ILs have emerged as powerful solvents for dissolving and fractionating lignocellulosic biomass. They can selectively extract lignin and cellulose, crucial components of plant matter, facilitating the development of sustainable bio-based chemicals and fuels. The ability of some choline-based ILs to dissolve keratin offers further potential in processing animal-based biomass for valuable products (Ovejero-Pérez *et al.*, 2021).

- Green Solvents for Organic Synthesis: The tunability of choline based ILs makes them excellent candidates to replace traditional volatile organic solvents in chemical reactions. They can promote various transformations, act as catalysts, and improve reaction efficiency while minimizing environmental impact. Their use has been demonstrated in polymerization, biomass conversion, and organic synthesis reactions (Gadilohar and Shankarling, 2017).

- Gas Absorption and Separation: The inherent binding capabilities of certain choline-based ILs, particularly with functional groups like b-diketonate, enable their use in selectively capturing gases like ammonia (NH_3) and carbon dioxide (CO_2). This property has implications for environmental remediation and the development of advanced gas separation technologies (X. Song *et al.*, 2023).

- Pharmaceuticals and Therapeutics: The biocompatibility and low toxicity of choline-based ILs create exciting opportunities in drug delivery, the development of novel pharmaceuticals, and biomedical applications. Their use as photosensitizers in photodynamic therapy (PDT) for cancer treatment, and their ability to stabilize nanoparticles for targeted drug delivery are promising areas of exploration (Demberelnyamba, Ariunaa and Shim, 2008; Chen *et al.*, 2024).

- Lubricants: Recent research has shown that choline based ILs can be directly utilized as high-performance lubricants without the need for additional additives. Their exceptional temperature and pressure tolerance make them attractive for industrial applications where traditional lubricants may degrade or fail (Mu *et al.*, 2016; Zhou and Qu, 2017).
- Advanced Materials: The ability of choline based ILs to dissolve metal oxides and enable electrodeposition paves the way for their use in functional materials synthesis and coatings. They could play a role in producing advanced materials for electronics, energy storage, and catalysis (Armand *et al.*, 2010; Gadilohar and Shankarling, 2017).

The potential applications of choline based ionic liquids are truly vast. Ongoing research efforts are focused on tailoring their structure and properties for even greater specificity and performance in various fields. Exploring their use in emerging technologies like 3D printing (Hossain *et al.*, 2023) and additive manufacturing (Nulwala, Mirjafari and Zhou, 2018) could lead to innovative material design and fabrication techniques. Furthermore, combining choline based ILs with nanoparticles or other functional materials promises the creation of novel composite materials with enhanced properties (Mohammed, Hadrawi and Kianfar, 2023). As research continues to uncover the full potential of these sustainable solvents, we can expect a growing number of applications across industries, contributing to technological advancements and a greener future.

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Appendix

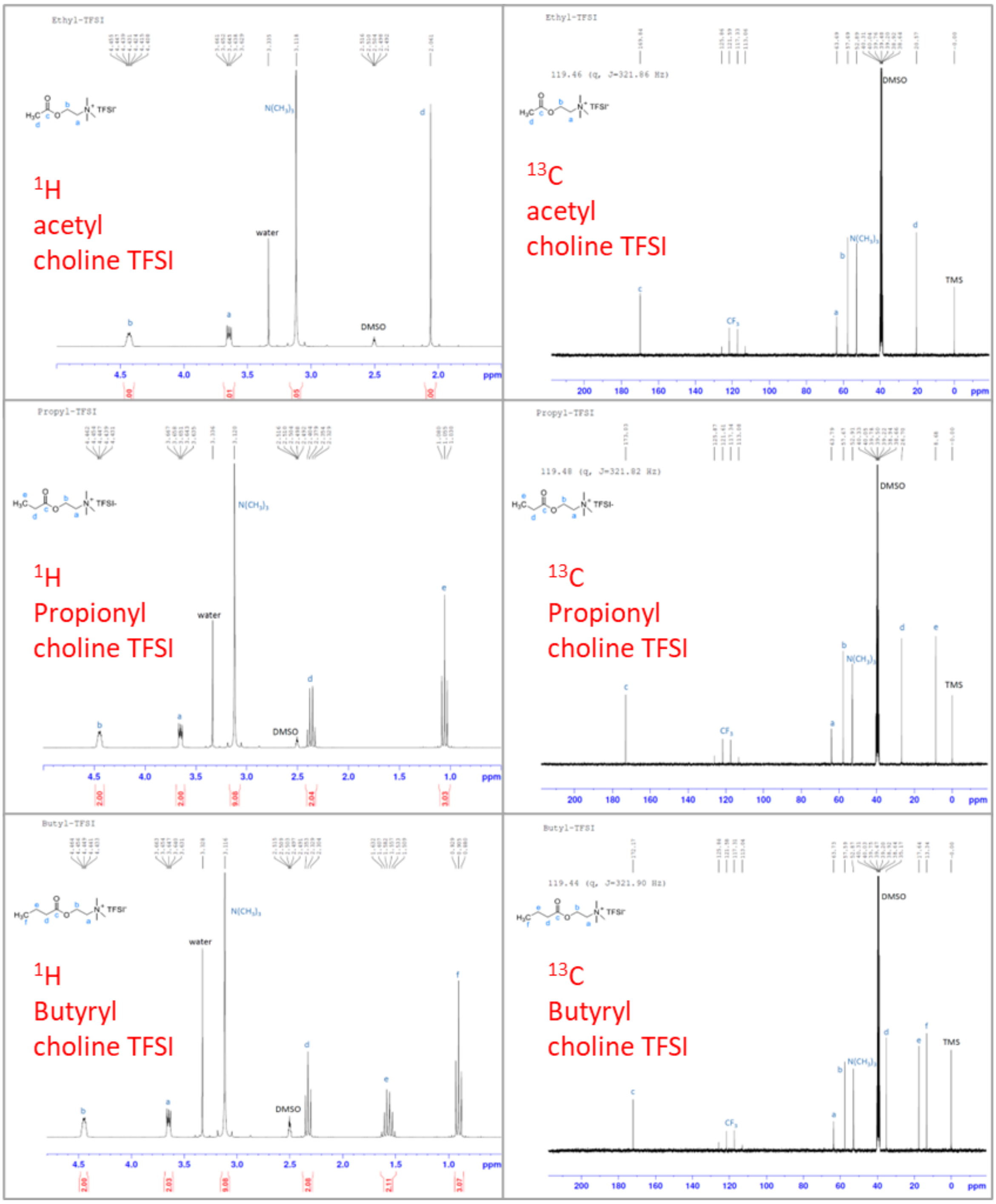

Figure S1. Proton and Carbon NMR spectra of choline ester ILs.