Electrolytes, separators and binders in lithium-ion batteries

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CONTENTS

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EXECUTIVE SUMMARY

Electrolyte formulations, separators and binders, previously considered to be 'passive' components, are now recognised as key components essential to support the safe and stable cycling of high-energy battery materials.

These high-energy battery materials – which include high-Ni NMC and Li metal – are widely accepted as being the next step change required for longer range EV vehicles, drones and aerospace applications. As the market for Li batteries increases, so too will the market for next-generation materials necessary to achieve the required performance. Existing companies that currently supply electrolytes, separator materials and binders stand to benefit from materials developments that improve cell performance. Moreover, there is also a great opportunity for new players to enter the market as new products will be necessary to address the requirements of next-generation batteries, such as higher temperature performance, greater sustainability and improved recycling capability. This report summarises the state of the art in electrolyte systems and discloses some of the current gaps that need to be addressed for improvement in energy density, safety and sustainability.
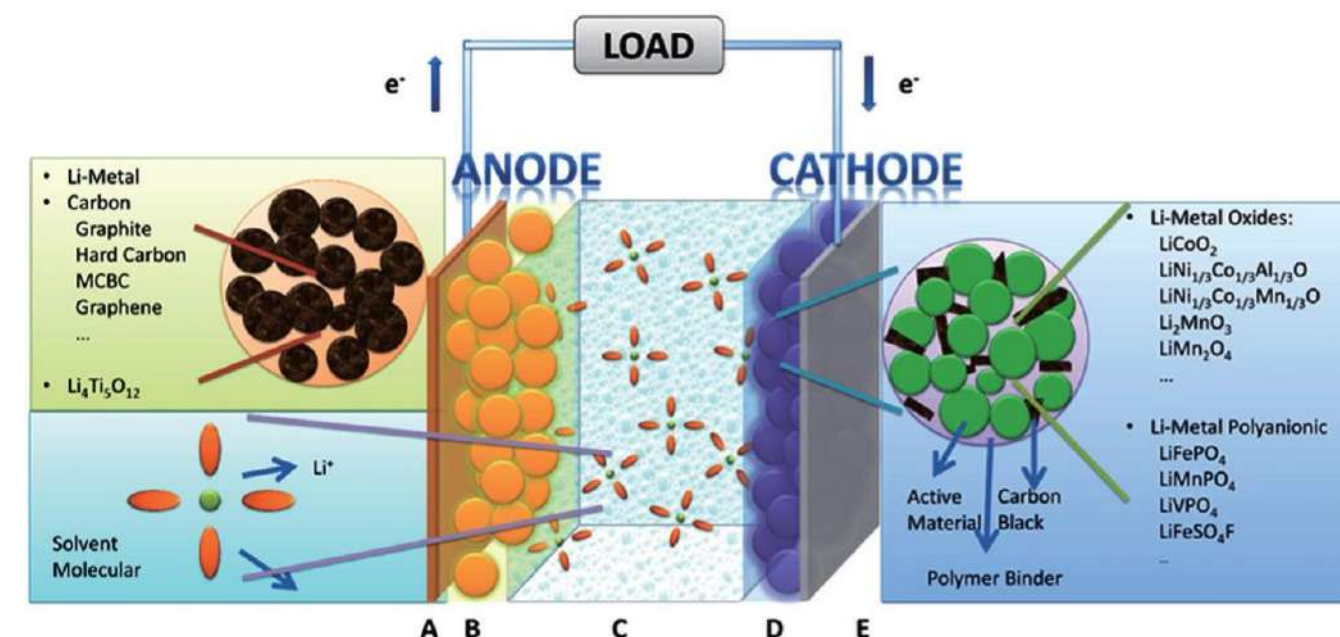
Figure 1. The configuration of a lithium battery.[2] The key components are Cu foil anode current collector usually (A), anode (B), electrolyte including separator (C), cathode (D) and Al foil cathode current collector (E).

1. INTRODUCTION

Li-ion batteries play a crucial role in our technology-driven society by powering a variety of devices including laptops, mobile phones, drones, electric vehicles and grid-scale energy storage systems. The rapid development of portable technologies demands compact and smaller batteries with higher energy densities, longer calendar lives, faster charging capabilities, improved safety operation across a wide temperature range, and at a lower cost. However, the current Li-ion battery technology is gradually approaching its practical energy density limit of ~300 Wh.kg^{-1}, and its intrinsic limitations hinder further improvements required to meet the exponentially increasing demands in commercial devices.[1] Therefore, new high-energy battery chemistries incorporating high-Ni NMC and Li metal electrode materials are required to develop next-generation Li batteries that can meet the ever-growing global energy demands. However, current conventional electrolyte systems do not readily support these requirements. Electrolyte formulations, separators and binders are now recognised as key components essential to support the safe and stable cycling of high-energy materials such as high-Ni NMC and Li metal.

In liquid electrolytes, the constituents include the Li salt, solvents and additives. However, traditional electrolytes are severely limited in high-temperature and high-voltage applications due to their poor thermal stability and electrolyte decomposition.[3] This necessitates the development of new electrolyte formulations that are suitable for the advanced electrodes required for use in high-energy density applications. However, the deployment of new electrolyte formulation chemistries has been hindered by the commercial success of traditional formulations for conventional Li-ion chemistries and the cost of incorporating high-performing systems. A lack of material knowledge and the understanding required to formulate the electrolytes for advanced electrodes are also significant contributors. Therefore, it is vital to develop approaches that provide a direct pathway for upscaling and access to expanded markets that will help to demonstrate new applications and benefit from economies of scale.

In addition to electrolytes, the separator is a key consideration for the safe and practical implementation of batteries.[4] Polyolefin-based polymeric separators are widely used in commercial Li-ion battery chemistries.[5] However, high temperatures (>100 °C) can induce them to shrink, melt and bring electrodes into contact, initiating thermal runaway resulting in the possibility of fires or explosions.[3, 6] High-purity alumina (HPA) coatings are now being adopted by the separator industry to improve the mechanical stability of polyolefin separators across a wide range of temperatures. These coatings also enhance the separator wettability,[7] which is essential for both commercial and novel electrolyte formulations in Li battery chemistries. The separator wettability also affects ion transport within the battery, ultimately influencing electrode performance and energy output. However, the influence of novel alumina processing methods on particle morphology and purity, and the resulting impact on cell performance, are not yet understood. A solution to overcoming issues associated with impurity leaching into the electrolyte would allow cheaper or alternative alumina processing methods to be established for the development of safer battery separators.

Binders are inactive materials used in small quantities in battery coating preparation to act as an effective dispersion agent to hold the active particles together and steadily adhere the coating to the substrate surface. The limitations with binder flexibility, swollen nature and degradation at elevated operating temperatures, require the development of more suitable binder materials for Li battery technologies.[8] Si-C composite electrodes are an emerging high-energy density anode material, however, the volume expansion of Si during cycling necessitates the use of flexible binders that can accommodate mechanical stresses and strains and promote longer cycle lifetimes.[9] Further, the operation of graphite electrodes at high temperatures can often be problematic due to the chemistry of the interphase layer that forms on the electrode surface. New electrolyte formulations are required to allow high-temperature operation while minimising performance degradation. Binder materials compatible with new electrolyte formulations are also essential for longer cycling Li battery chemistries. The long-term recyclability and environmental impacts are also important factors in developing binder materials.

In this report the electrolyte, separator and binder materials are evaluated for their existing and new market opportunities through product development and significant improvements in battery performance, benefits on the battery materials value chain, and their performance in current state-of-the-art Li battery chemistries. The development of new electrolyte systems will enable high-performance, high-energy-density Li batteries that can operate safely in extreme environments, such as at high temperatures and pressures, further expanding the market opportunities for Australian and global resources.

2. ELECTROLYTE SYSTEMS IN BATTERY VALUE CHAIN

2.1. Electrolytes

The electrolyte is a critical component in batteries that directly influences ion transport between the electrodes and the formation of the solid electrolyte interphase (SEI). The composition and structure of the SEI, which is dependent on the choice of electrolyte and the operating parameters, governs the electrode cycling stability, efficiency, capacity retention, safety, and cycle life.[40] Traditional carbonate electrolytes in Li-ion batteries often use lithium hexafluorophosphate (LiPF6) as the lithium salt, which is produced using processed lithium compounds (lithium carbonate and lithium hydroxide) derived from lithium spodumene raw material.[26] Currently, Australia has the world's largest lithium spodumene production.[26] The strong forecast growth in demand for Li-ion batteries will substantially benefit Australian miners and material producers, increasing profitability while generating a significant number of jobs in the battery materials manufacturing sector.

The limitations of conventional carbonate electrolytes, with respect to their electrochemical stability with high voltage cathode materials, [41] reactivity with high energy density Li metal anode materials [42] and the safety concerns due to their flammable nature, [43] have necessitated the development of novel electrolyte formulations. For instance, ionic liquid electrolytes, with their high electrochemical stability and good safety properties, or their hybrid formulations with low viscosity carbonate solvents, will create new market opportunities for high-performing battery chemistries.

The successful development of electrolyte formulations with leading-edge cell performance will benefit manufacturers with increased profitability of cell manufacture, lower lifetime cell cost and improved operational efficiency for end-users. Therefore, the benefits of electrolyte development will extend across the battery value chain.

2.2. High purity alumina (HPA)

Adding HPA particles to the LIB components may cause some new complexities for industrial processing. However, it offers various advantages such as higher thermal and mechanical stability, and safer devices with longer lifetimes. The purity of the alumina used in battery components – such as the cathode active material, the separator and the anode – all play a crucial role in battery performance.[44] In the case of composite separators, such as HPA-coated separators and porous membranes blended with HPA fillers, any impurities in the alumina particles, such as metal and non-metallic foreign particles, can cause defects that may damage the separator membrane and compromise cell performance by inducing local hot spots or short circuits.[45] With a separator thickness of only 20-25 μm, any small contamination with external particles can have devastating consequences. Therefore, the manufacturing process becomes significantly critical, especially when a high energy density cell is required. The employed alumina must be pure enough (typically 99.99% purity, metal impurities are less than a few ppm) to impart the mentioned benefits, otherwise it will be problematic and detrimental to battery performance.[46]

2.3. Binders

Binders make up only a small percentage of the overall mass and cost of a battery cell.[47] However, the cost of processing them needs to be considered. This is another area in which water-based systems are superior to the commonly used polyvinylidene fluoride (PVDF), as the water used to process them is considerably cheaper than the organic solvents (like N-methylpyrrolidone or NMP) used to process PVDF. Additionally, water-based systems are more environmentally friendly. Not only is water cheaper in itself, but it also cuts down the cost of several processing steps, such as drying of the processed electrodes due to the lower solvent boiling point.[48]

Another area in which binders play a role in reducing costs is recycling. With the projected increase in the use of LIBs, it is important to consider what will happen to the expended cells at the end of their life. This is important both practically – as LIB products contain critical minerals such as cobalt, nickel, copper and lithium – and ideologically, as battery technology is supposed to be an important component of a more sustainable future. With this in mind, it is clear that great effort should be made to recycle as much of the battery components as possible. This is especially true in the EV industry, which is showing high growth and uses large volumes of LIBs.[49] Great strides have been made in recycling efforts, but the way LIBs are constructed and the variety of materials they contain make it difficult. Multiple steps and harsh conditions, such as combustion at high temperatures and leaching with potent acids, are required.[50] Since the binder is the material that holds all the components of the electrode together, it presents a barrier to reclaiming the valuable minerals in the active materials. Even after high-temperature treatment, PVDF can still contaminate reclaimed material.[51]

Developing binders that degrade under milder conditions, while maintaining thermal and electrochemical stability during use, would simplify the recycling process and make it more economically viable to use recycled material in the manufacturing of new battery cells.

3. APPLICATION OF ELECTROLYTE SYSTEMS IN ENERGY STORAGE

3.1 Electrolytes

Traditional carbonate electrolytes

Most compositions of the organic liquid electrolytes for lithium-ion batteries (LIBs) contain $LiPF_6$ dissolved in a mixture of carbonate solvents. The most used carbonate solvents include cyclic and linear carbonates such as ethylene carbonate (EC), propylene carbonate (PC), dimethyl carbonate (DMC), ethyl methyl carbonate (EMC), diethyl carbonate (DEC). These cyclic diesters gained attention as electrolyte solvents for LIBs through their role in solid electrolyte interphase (SEI) formation on carbonaceous anodes. In single solvent electrolytes such as tetrahydrofuran (THF) or ethers, a coulombic efficiency ranging from 88% to 98% has been observed.[64–67] This was attributed to their oxidative decomposition on the cathode surfaces.[65, 66, 68] EC was then considered as a co-solvent and was found to play a key role in the formation of an effective SEI on graphite electrodes.[68,69] Electrolyte formulations composed of mixtures of DMC and EC have also shown high ionic conductivity and electrochemical stability.[70,71] Other linear carbonates such as DMC, EMC, and propylmethyl carbonate (PMC) are also being used with EC.[72, 73] Additives such as VC and fluoroethylene carbonate (FEC) have been incorporated to facilitate stable cathode SEI formation and suppress Li dendrite formation and transition metal dissolution from cathode materials.[74–76] Table 1 provides a summary of selected recent battery literature that uses carbonate electrolytes.

Room temperature ionic liquid (RTIL) electrolytes

Ionic liquids (IL) are a safer alternative to traditional carbonate electrolytes due to their superior chemical and electrochemical stability, negligible vapour pressure and nonflammability.[52] These properties of IL electrolytes have enabled the use of high energy density Li metal anodes, which are typically reactive with traditional carbonate solvents, as well as high voltage cathode materials (e.g., NMC, LMO) for the development of next-generation battery chemistries.

Table 1 presents selected IL electrolyte studies summarising their performance in various battery chemistries. Some of the more commonly studied ILs for Li as well as other metal (e.g., Zn, Na) battery chemistries consist of imidazolium, pyrrolidinium, and phosphonium IL cations with fluorinated anions (bis(fluorosulfonyl)imide [FSI]– and bis(trifluoromethanesulfonyl)imide [TFSI]–).[52] The composition of the electrolyte, including the type of IL cation and anion, Li salt and its concentration, will have a direct impact on the formation of the solid electrolyte interphase, ion diffusion, and ultimately battery performance.[53–56]

The solid electrolyte interphase is an electrically insulating but ionically conducting passivation film formed on the electrode surface through electrolyte decomposition and parasitic reactions that occur during battery cycling. As the SEI formation consumes the electrolyte and the electrode (e.g., Li metal) irreversibly, it is essential to facilitate the formation of a stable, homogeneous and efficient SEI, which will prevent further electrolyte decomposition and enable long-term battery cycling. The use of fluorinated anions in the IL electrolyte, such as [FSI] and [TFSI], has previously led to favourable SEI compositions and properties in Li metal battery chemistries.[53,55,57] Additives are also often employed in battery electrolyte formulations to modify the SEI, generally by sacrificing them during the initial cycling. Various additives, including solvents (e.g., n-pentane, vinylene carbonate, fluoroethylene carbonate) and salts (e.g., lithium bis(oxalato)borate, LiPF6) have been used in both IL and traditional electrolytes.[58–61]

However, ionic liquids are viscous compared to traditional carbonate electrolytes, and therefore, it is challenging to achieve high battery performance at room temperature. One of the ways to overcome this issue is to mix IL electrolytes with a co-solvent to reduce its viscosity. These hybrid electrolyte formulations will still preserve the favourable qualities of ionic liquids while improving their ionic conductivity. Various co-solvents, including 1,2-dimethoxyethane (DME), vinylene carbonate (VC) and ethylene carbonate (EC), have been used in IL electrolytes with Li metal, lithium iron phosphate (LFP), and NMC cathode materials (Table 1).[62,63]

TABLE 1: SELECTED STUDIES UTILISING TRADITIONAL CARBONATE AND IL BASED ELECTROLYTES FOR LI BATTERY CHEMISTRIES.

Cell Chemistry	Electrolyte	C rate or mA cm-2	Temp. (°C)	Cycles	CE (%)	Capacity retention (%)	Ref.
Al_2O_3 coated NMC|Li	1 M $LiPF_6$ in EC/ EMC (1/1 wt%)	0.5C	25	100	—	94.6	77
NMC811| Graphite	1 M $LiPF_6$ in 3:7 w/w EC/EMC, 2%VC	C/2	25	500	100	98	78
Nd_2O_3 coated NCM811| Li	1 M $LiPF_6$ in EC: DMC: EMC (1:1:1 v%)	1C	25	200	—	88	79
Nb doped $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ | Li	1 M $LiPF_6$ in EC/DMC (1/1 v%)	5C	25	200	100	83.3	80
1 wt% Li_4MgWO_6 + NMC|Graphite	1 M $LiPF_6$ in EC/DMC (1/ 1 v/v)	1C	25	2000	—	89%	81
LTO coated $LiNi_{0.8}Co_{0.1}Mn_{0.1}O_2$ | Li	1 M $LiPF_6$ in EC/DMC (1/1 v%)	1C	25	170	99.75	75.86	82
4.6 mAh cm_{-2} NMC 622| Cu	3.2 m LiFSI in $[P_{1222}][FSI]$	1.75 mA cm_{-2}	50	100	99.4	53	83
3.5 mAh cm_{-2} LFP| Li	3.2 LiFSI in 80[C_3mpyr][FSI] +20DME	1 mA cm_{-2}	50	350	99.8	—	84
2.8 mg cm_{-2} NMC 88| Li	20 mol% LiTFSI in [C_4mpyr][FSI]	0.3C	20	1000	99.94	88	56
2-3 mg cm_{-2} NMC 111|Sn-C	20 mol% LiTFSI in[C_4mpyr][TFSI]	25 mA g_{-1}	40	500	98	99	85
4.03 mAh cm_2 NMC 622 single crystal| Li	3.2 m LiFSI in $[P_{1222}][FSI]$	1.75 mA cm_{-2}	50	100	99.91	80.6 (50th cycle)	54
LFP| Sn-C	0.2 m LiTFSI in [C_4mpyr][FSI]	0.12 mA cm_{-2}	40	2000	99.9	98	85

3.2 HPA

LIB manufacturing techniques become significantly critical as the energy density of the battery increases. To tackle the emerging safety and long-term efficiency issues of LIBs, various techniques have been investigated. Among these is the use of ceramic powders to enhance SEI chemistry, limit the electrode expansion and improve the thermal and mechanical stability of the cell components. HPA is one of the most widely accepted ceramic materials due to its unique properties, including its inertness to all the chemicals present in LIB components, high thermal stability, mechanical robustness and availability. These properties make HPA ideal for various applications as an additive material in electrodes or separator coating to improve the thermal and mechanical stability of the separator and avoid detrimental contraction and dendrite growth.

Coating a PP/PE (polypropylene/polyethylene) separator using HPA was invented by Celgard in 2000; since then, multiple investigations have been done in various battery systems in which the presence of HPA demonstrates a significant improvement in physical and chemical properties of this porous membrane. Moreover, the HPA-coated separator has been shown to prevent dendrite puncture and separator shrinkage during Li insertion, as well as preventing over-heating, enhancing cell safety. Some of the more recent cell performance improvements associated with the use of HPA in cell components, which was incorporated via various techniques, are listed in Table 2.

3.3 Binders

PVDF has been the most used binder material in LIB manufactures. Much of the recent literature has focused on finding more environmentally friendly alternatives that also show better performance. Carboxy methyl cellulose (CMC) and polyacrylic acid (PAA) are the two materials that have gained the most attention in this area. Additionally, the chemically diverse nature of binders has been exploited to produce specialized materials, like binders that are themselves conductive. An issue that has been particularly focused on is the expansion of Si particles during lithiation in Si anodes. By changing the chemistry of the binder and making it, for example, crosslinked or self-healing, this challenge could be overcome. This example illustrates the power of using diverse binders to overcome challenges presented by certain systems. Table 3 summarises a few characteristics currently being investigated in binder materials that have been shown to have certain advantages over current materials. Naturally, there is often overlap between these characteristics, with for example CMC being naturally sourced and water based. It is also worth pointing out the role of powerful material science techniques in investigating these research problems. Techniques like Scanning Electron Microscopy (SEM), X-Ray Photoelectron Spectroscopy (XPS), and Time of Flight Secondary Ion Mass Spectrometry (ToF-SIMS) make it possible to visualize the changes and track the elemental distribution of the various components within the electrodes before and after use.

TABLE 2: RECENT STUDIES USING ALUMINA COATED SEPARATORS AND ELECTRODE MATERIALS.

Technique	Advantages	Ref.
HPA coated NMC cathode	• Stability and cyclability improvement of the cathode material • Higher capacity retention and delayed active material pulverisation • Prevention of metal dissolution and cross-talk	86–88
HPA coated PP/PE separators	• Enhanced thermal conductivity and effective heat dissipation and stability • Li+ transport, ionic conductivity and wettability were improved • Lower internal resistance and less self-discharge • Enhanced dimensional and mechanical stability up to 140 °C	7,89–92
HPA coated graphite	• The risk of Li metal deposition throughout cycling under harsh environment is sensibly less • Higher mechanical stability • Improved SEI structure and less dead Li formation on the surface	93
HPA coated Li metal anode	• Improved cycling performance • Homogeneous Li nucleation and growth • Li expansion over cycling was limited • Prevention of the internal resistance build-up over cycling Formation of more conductive SEI layer by formation of Li-Al-O and Al-F bonds • Well-regulated Li deposition and dendrite-free	94,95

TABLE 3: CHARACTERISTICS CURRENTLY BEING INVESTIGATED IN BINDER MATERIALS.

Characteristic	Advantages	Ref.
Renewable source	• More environmentally friendly	33,97–101
Water-based	• More environmentally friendly • Cheaper	33,97–105
Chemical interaction with active material	• Stronger adhesion • Enhanced electrode integrity • More consistent cycling performance	105–107
Negatively charged groups	• Selective intercalation of Li ions while cointercalation of solvent is suppressed	108
Conductivity	• Enhances overall conductivity	38,109
Crosslinking	• Enhanced electrode integrity • More consistent cycling performance	37,110,111
Self-healing	• Enhanced electrode integrity as cracks that form during operation are healed • More consistent cycling performance	39,112,113

4. GLOBAL AND AUSTRALIAN MARKET OPPORTUNITIES, CAPABILITIES AND BARRIERS FOR ELECTROLYTE SYSTEMS

4.1 Electrolytes

As shown in Figure 1, all types of Li-ion batteries (LIBs) are comprised of two electrodes (anode and cathode) separated by a liquid or solid electrolyte. During the battery charging and discharging processes, the electrolyte enables the Li-ions to move between the electrodes to store or produce energy. In a rechargeable battery, this is a reversible process that enables multiple charge/discharge cycles. Therefore, it is necessary for electrolytes to have high ionic conductivity and superior chemical and electrochemical stability. Since the introduction of Li-ion batteries to the commercial market in the 1990s, their global market size was valued at US$53.6 billion in 2020 and was expected to grow at a 19.0% CAGR (compound annual growth rate) until 2028[10].
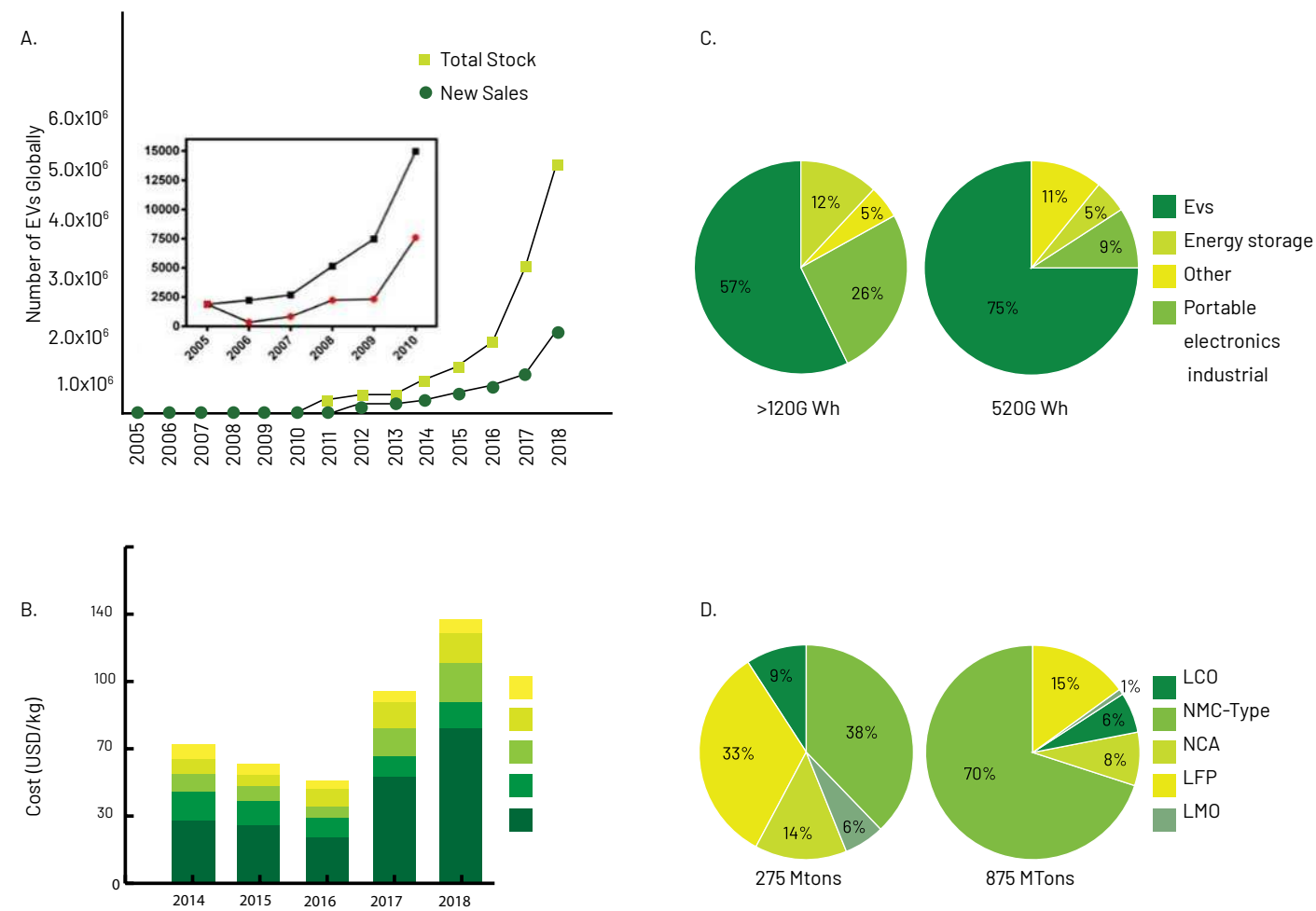

Figure 2. (A) Global historical EV sales and (B) the price of valuable metals used in LIBs. Projection of LIB market share based on (C) application and (D) cathode composition assuming that Tesla, Inc remains the only major EV manufacturer to adopt NCA cells in 2025.[11]

This constant growth of technological advancements increases the scope of battery electrolyte applications. In 2020, the global battery electrolyte market size was valued at US$4.9 billion, and it is expected to reach US$8.9 billion by 2028 with a CAGR of 7.7% during the forecast period.[12] Some of the key factors driving the growth of the battery electrolyte market result from the increasing government initiatives across the world that aim to reduce the global carbon emission by adopting electric vehicles (EVs) and renewable energy storage systems (ESS). In addition, the rapid production, and sales of consumer electronic devices, such as laptops, tablets, smartphones, and fitness devices, are expected to create a positive impact on the global electrolyte market.

One of the ongoing barriers to the large-scale deployment of the electrolyte market is the high initial capital cost of Li-ion batteries in major end-user applications, transport and stationary ESS sectors[13]. In EVs, the upfront and total costs for the vehicle ownership are significantly higher than for an internal combustion engine vehicle, and more than 50% of the EV cost is attributed to the battery pack.[14] The electrolyte component currently contributes to approximately 5% of the EV battery pack cost.[13] Similarly, while the uptake of photovoltaics in industrial and household settings is high, their integration into a battery ESS is limited due to the high investment cost. The safety concerns raised by recent fire incidents with EVs and portable electronic devices also have a negative impact on the traditional electrolyte market due to their flammable nature in Li-ion batteries.[15, 16] The traditional carbonate electrolyte market is also challenged by the demand for new electrolyte formulations that are suitable for the rapidly evolving end-user requirements (e.g., high-temperature applications, high energy density) and the deployment of other technologies in the ESS (e.g., residential Li-ion battery packs).

With increasing opportunities in the research sector, ionic liquids (ILs) are considered strong candidates for the next generation of electrolytes to overcome the safety issues associated with traditional electrolytes and for the development of high-energy density battery technologies. Global electrolyte manufacturing company Solvionic and the Australian company Boron Molecular are some of the companies actively pursuing the IL production path. However, to reduce the cost of IL electrolytes and their highly viscous nature compared to carbonate solvents, hybrid electrolyte formulations are being screened by combining the beneficial properties of both ILs (high electrochemical stability, safety) with traditional carbonate solvents (low viscosity). This will create new market opportunities for the current key electrolyte manufacturers, such as Mitsubishi Chemical Corporation (Japan), Ube Industries (Japan), Shenzhen Capchem Technology Co., Ltd. (China), Johnson Controls (US), 3M (US), BASF SE (Germany), LG Chem (South Korea), and American Elements (US), to expand their market footprint. Australia is currently at the beginning of the path for the battery manufacturing supply chain with the competitive advantage of mineral sources. With government initiatives on the battery industry's implementation and growth, and the adoption of EVs, further progress is expected in the local battery electrolyte manufacturing capabilities.

4.2 HPA

The global HPA market was valued at US$1.3 billion in 2019 and is predicted to reach US$4.8 billion by 2026 for various applications.[17, 18] The demand for HPA is increasing in LIB manufacturing as a result of the environmental regulations imposed in various countries such as the USA, Germany, India and China to reduce CO2 emissions and decrease the global dependency on conventional fuels. One of the main applications of HPA in LIBs is in coating separators, which is an area that is expected to significantly grow over the next decade, especially for portable electronic devices, electric vehicles, and stationary applications in which high-capacity large-sized cells are required. For instance, the usage of separators for the LIB storage systems in Europe indicates revenues of US$1.3 billion for 2016 with an expected CAGR of 13% annually and a projection of US$3.2 billion for 2025.[19] Moreover, the total LIB market in 2016 reportedly consumed about 1200 million m^2 of separators, which demonstrates a tremendous global market opportunity in this field.

Commercialised separators are usually developed for a specific type of battery, therefore, there are about 100 companies manufacturing various types of separators present in the alternative energy market. 20 Some of the major separator suppliers are Celgard LLC, Asahi Kasei, Entek Membranes LLC, BASF, Evonik Industries AG, Exxon Mobil Corporation/TonenGeneral Sekiyu K.K, Chemicals Corporation, SK Energy, Hitachi Maxell Global, LG Chem, Porous Power Technologies and UBE Industries.[20, 21]

To date, microporous polymeric-based membranes filled with liquid electrolytes have been predominantly commercially employed for LIBs. However, their performance, energy density and safety have been limited, especially for applications that require heavy-duty batteries that can perform over a wide range of temperatures and can deliver higher energy density. Therefore, ceramic-coated separators for secondary batteries have gained significant interest as an approach to potentially overcome the aforementioned challenges and manufacture safe and reliable separators with higher efficiency.[21–23]

4.3 Binders

Although they often do not receive as much attention as other LIB parts, binders are crucial as they hold together all other components of the electrodes, i.e., the active and conductive materials. To ensure conductivity, all electrode components need to be distributed homogeneously.[24] During manufacturing, the binder's physical properties, such as its viscosity, influence the processability and ultimately the composition of the electrodes.[25]

Australia is well-positioned to benefit from rising LIB demand as society transitions towards renewable energy, however, it could be taking further steps to strengthen its global position.[26] All the major raw materials needed for the manufacture of LIBs (lithium, nickel, manganese, cobalt, HPA, etc.) are present in Australia in large mineral deposits and are currently exported to varying degrees. These materials are then readily refined into battery-grade chemicals and used in cell manufacture, but this is rarely done within Australia.[27] If Australia is to take further advantage of its position, move up the value chain, and establish a battery industry, binders are going to be a part of that equation. While it might not be economically viable for Australia to compete with cheaper jurisdictions in the manufacture of traditional, established chemistries, existing technical expertise could be leveraged to enter more niche markets.

The global market for binders in lithium-ion batteries is forecast to grow at a CAGR of 24% during the period between 2021 and 2025.[28] Unlike in the active material market, where multiple different chemistries take up a large share of the market, the binder market has been largely dominated by PVDF, which offers good processability and electrochemical stability.[29] It does, however, have significant drawbacks as well.[27] For example, as an insulating material, PVDF necessitates the addition of conductive additives to the electrodes in order to ensure continuous conductivity.[30] The use of PVDF also comes with environmental concerns, including the need for the use of toxic solvents such as NMP in its processing and the release of harmful, fluorinated degradation products when burned as part of the recycling process.[31–33]

Many companies operating in the battery binder market, such as Arkema (France), Solvay (Belgium) and Kureha (Japan), focus on producing PVDF. Other companies like Ashland (USA), DuPont (USA), JSR (Japan) and Zeon Specialty Materials (Japan) are focusing on water-based binder systems. As mentioned, Australia's battery industry is still underdeveloped, so there are only a few small examples of companies producing binders, such as AnteoTech with its crosslinked system for silicon anodes.

The move to replace PVDF with water-based and often naturally-sourced binder systems like CMC, PAA and sodium-alginate (Na-ALG) is one of the main opportunities in the binder market. These materials are more environmentally friendly, cheaper, and contribute to better battery performance due to their higher degree of interaction with the active material, preventing cracking after extended operation. This is especially important in Si anodes, which show superior performance compared to standard graphite anodes but undergo drastic size changes upon alloying with lithium. A binder that demonstrates greater adhesion with the Si particles due to its chemical makeup will accommodate this change better without compromising the integrity of the electrode. In general, binders deserve more focus because of the advantage of their chemical variety. Considering their nature as polymers, it is much easier to introduce new functionality into them. For example, the binder network can be crosslinked to enhance its integrity, a polymer that is itself conductive can be used to enhance the overall conductivity, or self-healing properties can be introduced. By further exploiting this potential and innovating in this space, new niches in the battery market can be opened.

5. CONCLUSION

High energy density batteries are required to meet the ever-growing energy demands of our society. A strategy to meet these demands focuses on the search for high energy density electrodes such as high-Ni NMC and Li metal. However, this brings up a new set of limitations in different components of the battery, such as the electrolyte, binder and separators, to maintain the performance of current Li-ion battery technologies. For this reason, new chemistries for electrolytes, binders, and separators should be investigated that present thermal, chemical and electrochemical stability.

More specifically, new electrolyte formulations are required to realise new Li battery chemistries incorporating high-Ni NMC, Li metal, and Si-C composite electrode materials that can perform under extreme conditions, including potential natural disasters such as in mining environments, in the presence of flooding and fires. Ionic liquids (ILs) and hybrid electrolytes composed of ILs and co-solvents, as well as additives, are new types of electrolytes that can meet those requirements. Among the different families of ILs, short alkyl phosphonium and pyrrolidinium-based ILs are the ones mostly used in the literature for Li battery technologies. However, more research is still necessary to understand the reaction mechanism, SEI formation, and possible side reactions during battery operation, which will depend on the electrode and electrolyte chemistries.

On the other hand, HPA coatings are now being adopted by the separator industry to improve the mechanical stability of polyolefin separators across a wide range of temperatures. An additional benefit of using HPA is the enhancement in separator wettability, which enhances the ion transport between electrodes. However, the impact of alumina impurities is still unknown, requiring more research to clarify the influence on separator performance and, consequently, overall battery performance. Reducing the purity of the HPA will have a positive impact on reducing the cost of the binder.

Another component of Li-ion batteries that can be improved is the binder material, which holds together the active and conductive materials of the electrode. Recent research in this area has had two major themes. Firstly, it has explored the replacement of PVDF with more environmentally friendly, cheaper, and performance-enhancing water-based systems. Secondly, it has explored the exploitation of the chemically diverse nature of polymeric binders to introduce new functionality, such as crosslinking and self-healing, which also enhance the long-term performance of the battery cell.

6. GLOSSARY

CAGR	Compound Annual Growth Rate	NMC	Nickel Manganese Cobalt
CMC	Carboxy Methyl Cellulose	NMP	N-methylpyrrolidone
DEC	Diethyl Carbonate	PAA	Polyacrylic Acid
EC	Ethylene Carbonate	PC	Propylene Carbonate
DMC	Dimethyl Carbonate	PMC	Propyl Methyl Carbonate
EMC	Ethyl Methyl Carbonate	PVDF	Polyvinylidene Fluoride
EV	Electric Vehicle	SEI	Solid Electrolyte Interphase
ESS	Energy Storage System	VC	Vinylene Carbonate
FBICRC	Future Battery Industries Cooperative Research Centre	Wh	Watt hours
HPA	High Purity Alumina		
IL	Ionic Liquids		
LIB	Lithium Ion Battery		

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