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Recent Advances in Non-Flammable Electrolytes for Safer Lithium-Ion Batteries

Neha Chawla [1],* , Neelam Bharti [2] and Shailendra Singh [1]

[1] Environmental, Health and Safety, Carnegie Mellon University, Pittsburgh, PA 15213, USA;
shailen2@andrew.cmu.edu

[2] Mellon Institute Library, Carnegie Mellon University, Pittsburgh, PA 15213, USA;
nbharti@andrew.cmu.edu

* Correspondence: nehac@andrew.cmu.edu

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Abstract: Lithium-ion batteries are the most commonly used source of power for modern electronic devices. However, their safety became a topic of concern after reports of the devices catching fire due to battery failure. Making safer batteries is of utmost importance, and several researchers are trying to modify various aspects in the battery to make it safer without affecting the performance of the battery. Electrolytes are one of the most important parts of the battery since they are responsible for the conduction of ions between the electrodes. In this paper, we discuss the different non-flammable electrolytes that were developed recently for safer lithium-ion battery applications.

Keywords: lithium-ion battery; non-flammable; electrolyte; safety; battery fire

1. Introduction

Rechargeable lithium-ion (Li-ion) batteries are widely used in portable electronic devices and are considered as the most potential power source for electric vehicles due to their high energy density and long cycle life. A Li-ion battery consists of a positively charged cathode, negatively charged anode, separator, electrolyte, and positive and negative current collectors. While discharging, the lithium ions travel from the anode to the cathode through the electrolyte, thus generating an electric current, and, while charging the device, lithium ions are released by the cathode and then go back to the anode. Figure 1 shows the basic working principle of a Li-ion battery. Since the electrolyte is the key component in batteries, it affects the electro-chemical performance and safety of the batteries.
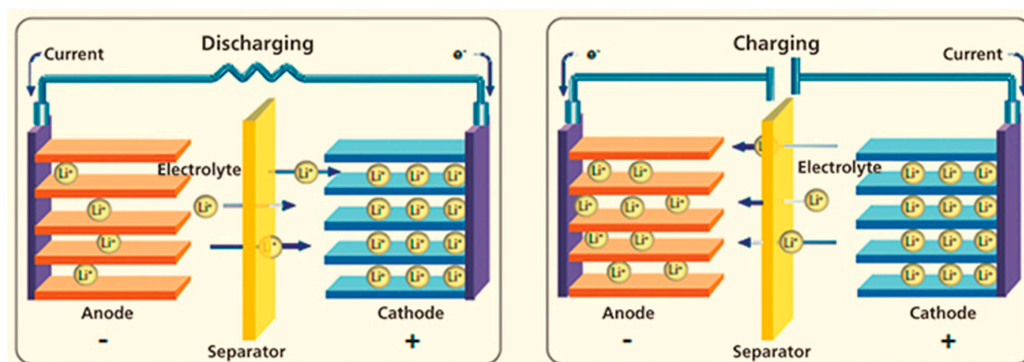

Figure 1. Basic working principle of a lithium-ion (Li-ion) battery [1].

The most common devices we use, such as cell phones and other common electronics, use Li-ion batteries. Recently, several accidents were reported due to Li-ion battery failure. In the past few years,

several cases were reported where cell phones, laptops, hoverboards, etc. caught fire due to Li-ion battery failure. According to a news report, there were 92 cases of Samsung Galaxy Note fires, 26 burns, and 55 property damages [2]. The Federal Aviation Administration (FAA) reported that the Li-ion battery accidents increased from 31 incidents in 2016 to 46 incidents in 2018 [3]. As per the FAA report, one Li-ion battery "incident" occurs every 10 days either on airplanes or at airports. In 2010, a United Postal Service (UPS) cargo plane crashed in the United Arab Emirates because of a fire caused by a shipment of Li-ion batteries in the cargo hold [4]. In November 2017, the Li-ion battery of a camera exploded at Orlando International Airport at a security checkpoint, which caused a terminal to be evacuated [5]. A Li-ion battery-containing device exploded and caught fire in a passenger suitcase loaded in the cargo on a Delta Connection/SkyWest flight [6]. A battery-operated vape pen caught fire during a Transportation Security Administration (TSA) X-ray screening at Denver International Airport in January 2018. In another incident, in January 2018, a lithium-battery power bank caught fire on a flight that was scheduled to depart to Shanghai. Recently, there were several reports of Tesla vehicles catching fire [7]. In June 2018, a battery pack of Tesla Model S caught fire resulting in a casualty [7]. In 2016, the same model also caught fire during a test drive event in France [8].

A battery under normal work conditions consists of organic solvents and lithium salt. Water and heat cause the thermal dissociation of $LiPF_6$, the produced Lewis acid PF_5 attacks the solvent molecules, and the combustion of large amounts of active free radicals leads to thermal runaway. Li-ion battery fires are typically a result of heat generated due to a short circuit within one or more of the battery's cells. Generated heat in the cell ignites the chemicals within the battery, which leads to "thermal runaway". Thermal runaway temperatures can go as high as 1000 °F, creating intense pressures inside the battery, leading to an explosion in the flammable liquid electrolyte. Figure 2 shows the schematic representation of a thermal runaway process [9].
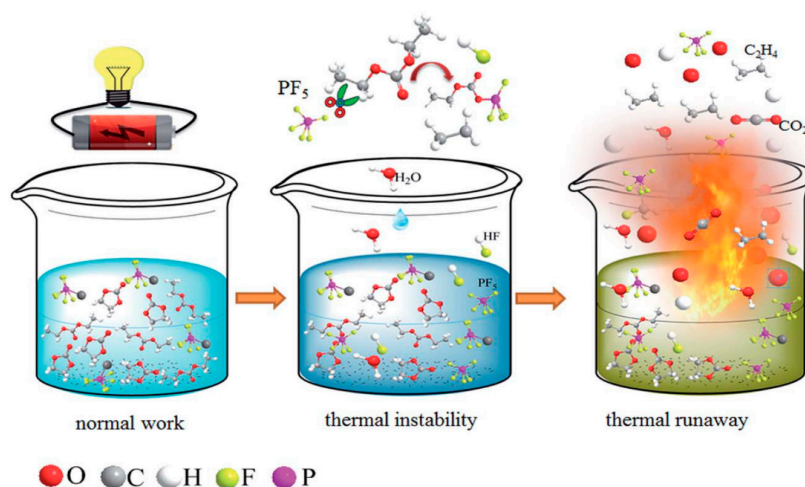

Figure 2. Schematic of the thermal runaway process of the Li-ion battery electrolyte [9]. Republished with permission of the Royal Society of Chemistry, from A self-cooling and flame-retardant electrolyte for safer lithium-ion batteries; Lihua Jiang, Qingsong Wang, Ke Li, Ping Ping, Lin Jianga, and Jinhua Sun; 2, and © 2018; permission conveyed through Copyright Clearance Center, Inc.

As the demand for Li-ion batteries (LIBs) is increasing, the safety concerns of the conventional carbonate-based electrolytes also increased in the commercial LIBs. Developing new electrolyte systems with improved safety features is one of the high priorities being investigated by several researchers. Conventional electrolytes are mostly composed of lithium hexafluorophosphate ($LiPF_6$) dissolved in a mixture of ethylene carbonate (EC) and linear carbonates [3]. Carbonates have high volatility and flammability. However, the high flammability of linear carbonates, such as diethyl carbonate (DEC), dimethyl carbonate (DMC), and ethyl methyl carbonate (EMC), is the biggest safety concern of lithium-ion batteries [10]. To date, researchers made several efforts to formulate safe electrolytes with

enhanced battery performances [11–14]. In this review article, we compile literature and discuss a few recent advances in electrolyte formulation toward safe Li-ion batteries.

2. Results and Discussion

2.1. Organosilicon-Containing Electrolytes

Organosilicon compounds are compounds with carbon–silicon bonds. They receive considerable attention as electrolytic solvents for energy storage devices because of their low flammability, high thermal and electrochemical stability, and environmentally benign characteristics. Wang et al. synthesized two fluorosilanes with oligo(ethylene oxide) units through hydrosilylation of chlorosilane with allyl substituted oligo(ethylene oxide) ether, followed by fluorination with potassium fluoride, namely fluoro(3-(2-(2-methoxyethoxy)ethoxy)propyl) dimethylsilane ($MFSM_2$) and difluoro(3-(2-(2-methoxyethoxy)ethoxy)propyl) methylsilane ($DFSM_2$) [11]. The fluorination resulted in lowering the viscosity, and increasing the dielectric constant and oxidative potential as compared to their non-fluorinated counterparts. $DFSM_2$-doped electrolyte exhibited high voltage and improved thermal stability when used with a lithiated graphite anode and a delithiated $LiCoO_2$ cathode, making it a safe electrolyte for lithium-ion batteries. The electrolyte, 1 M $LiPF_6$ (lithium hexafluorophosphate) in EC/$DFSM_2$/EMC ($v/v/v$ = 2/3/5) (ethylene carbonate/difluoro(3-(2-(2-methoxyethoxy)ethoxy)propyl) methylsilane/ethyl methyl carbonate) with the addition of 5 wt.% fluoroethylene carbonate (FEC) displayed an improved voltage limit, which led to a cyclability performance with 92.5% capacity retention at a voltage cutoff of 4.4 V after 135 cycles in a $LiCoO_2$(LCO)/graphite full cell [11].

2.2. Ionic Liquid Electrolytes with/without Solvents

Ionic liquids are salts with poorly coordinated ions, resulting in these solvents being liquid below 100 °C, or even at room temperature. Ionic liquids received great attention recently for their use in batteries. Ionic liquids (ILs) have low volatility, are non-flammable, and have high electrochemical and thermal stabilities, due to which they are used for energy storage and conversion applications, such as super capacitors, solar cells, and batteries [15–17]. Three choline-based ionic liquids functionalized with trimethylsilyl, allyl, and cyanoethyl groups for high-voltage Li-ion batteries [12] were achieved via a two-step method that consisted of an anion exchange reaction of chlorine ion with LiTFSI (lithium bis (trimethylsulfonyl) imide) and a functionalization reaction with hexamethylsilazane, allyl bromide, and acrylonitrile [12]. The hybrid electrolyte was constructed by doping with 0.6 M $LiPF_6$/0.4 M lithium oxalydifluoroborate (LiODFB) as salts and dimethyl carbonate (DMC) as a co-solvent. Thermal properties of the electrolytes were found with the thermal decomposing temperatures in the order of AN1IL-TFSI (325 °C) < CEN1IL-TFSI (330 °C) < SN1IL-TFSI (336 °C) (AN1IL-TFSI (IL with allyl group), CEN1IL-TFSI (IL with cynoethyl group, SN1IL((2-trimethylsililoxyethyl) trimethylammonium bis(trifluoromethanesulf-onyl)imide).An $LiCoO_2$/graphite full cell using SN1IL/DMC (v/v = 1/1) doped with 0.6 M $LiPF_6$/0.4 M LiODFB (lithium oxalydifluoroborate) salts showed a cyclability of over 90 cycles with a capacity of 152 mAh·g^{-1}, a voltage cut-off of 4.4 V with good rate capability, retaining 72% capacity of 0.2C at the 2C rate. Only one-quarter of the propagation rate of the electrolyte was obtained as compared to commercial reference electrolyte (1 M $LiPF_6$ EC/DEC/DMC, $v/v/v$ = 1/1/1), which is a good safety feature.

To design and develop safer and high-energy-density Li-ion batteries, Moreno et al. developed an electrolyte comprising PYR13TFSI/PYR13FSI ionic liquid with LiTFSI salt, where PYR13 stands for *N*-methyl-*N*-propyl pyrrolidinium, TFSI is bis(trifluoromethanesulfonyl)imide, and FSI is bis(fluorosulfonyl) imide [18]. The electrolyte exhibited a good ionic conductivity even at −20 °C with an average electrochemical stability window of 5 V. Electrolytes were also developed by mixing ethylene carbonate (EC) with ionic liquids based on ringed ammonium cations with ring sizes of 7, 6, and 5, i.e., azepanium, piperidinium, and pyrrolidinium, respectively [13]. The designed

electrolytes had lower viscosities, improved conductivity, and better stability as compared to ionic liquids. Ionic liquids have good thermal stability, whereas EC is flammable; the electrolyte obtained by combining the two components was non-flammable and exhibited good thermal stability above 130 °C. Li/LiMn$_{1.5}$Ni$_{0.5}$O$_4$ (LMNO) and graphite/Li half batteries were fabricated with a fluoroethylene carbonate additive which led to discharge capacities of approximately 115 and 287 mAh·g^{-1} at Coulombic efficiencies of 95% and 99%, respectively, over 100 cycles at a rate of C/12.

Li-ion batteries were prepared and tested by Gao et al. with an inorganic non-aqueous liquid electrolyte/LiAlCl$_{4.3}$SO$_2$ (IE) and LiFePO$_4$ cathode [14]. The batteries displayed stable cycling, higher discharge capacity, and better performance as compared to batteries with organic electrolytes. The ionic conductivity of the electrolyte was found to be better than organic electrolyte and it exhibited better safety performance and non-flammability.

2.3. Flame-Retardant Solvents

Flame-retardant solvents generally compromise the battery performance [19–22]. To explore this idea, Wang et al. studied an organic electrolyte using a sodium or lithium salt and trimethyl phosphate (TMP) [23]. TMP was used as a solvent because it is identified as a good flame retardant and has high oxidative stability and low viscosity [23]. At the C/5 rate, they obtained 1000+ cycles (over one year) with almost zero degradation when cycling with carbon/graphite anodes for sodium-ion/Li-ion batteries. NaFSA/LiFSA (NaN(SO$_2$F)$_2$/LiN(SO$_2$F)$_2$) salts were used due to weak cation–anion interactions, which offer high ion transport even at elevated concentrations. Figure 3 shows the flame test of NaFSA/TMP. When used in a battery with a composition of 5 V class LiNi$_{0.5}$Mn$_{1.5}$O$_4$/graphite, the electrolyte exhibited a stable 100 cycles of discharge/charge at C/5. During the study, obtained Coulombic efficiency was 99.2%, thus indicating its potential application for safe and high-voltage Li-ion batteries.
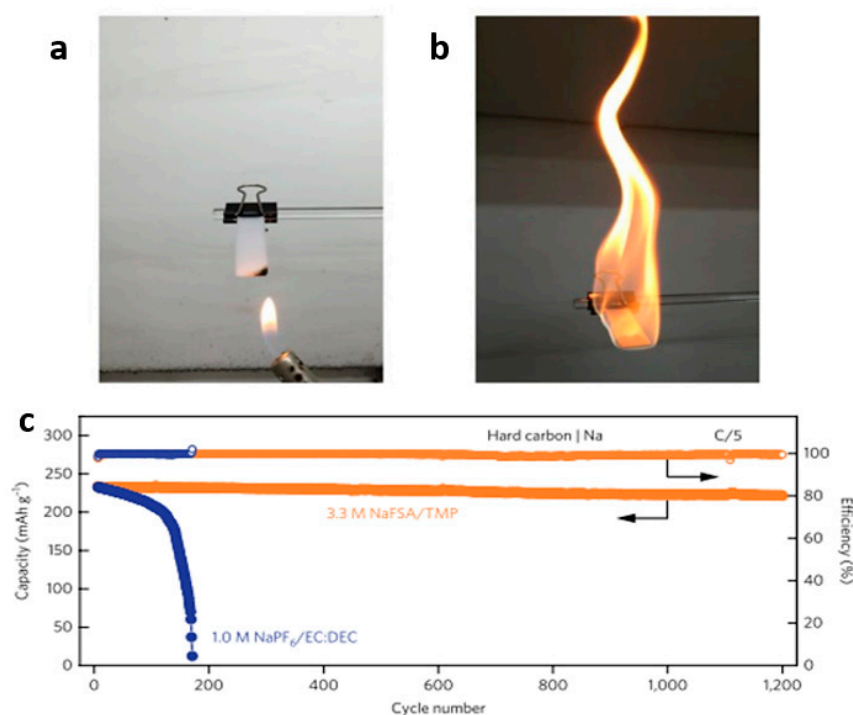

Figure 3. (**a,b**) Flame tests of laboratory-made NaFSA (NaN(SO$_2$F)$_2$)/trimethyl phosphate (TMP) electrolyte and conventional 1 M NaPF$_6$/EC:DEC (*v/v* = 1/1) electrolyte [23]. (**c**) cycling performance and Coulombic efficiency of half cells using concentrated 3.3 M NaFSA/TMP electrolyte and conventional 1 M NaPF$_6$/EC:DEC (*v/v* = 1/1) electrolyte. Reprinted with permission from Springer Nature, Nature Energy; Fire-extinguishing organic electrolytes for safe batteries, Jianhui Wang, Yuki Yamada, Keitaro Sodeyama, Eriko Watanabe, Koji Takada, Yoshitaka Tateyama, and Atsuo Yamada, © 2018.

Adiponitrile (ADN) is considered as a safe electrolyte because it is chemically stable and has a high boiling and flash point and low vapor pressure. ADN was used as a single-electrolyte solvent with lithium bis(trimethylsulfonyl)imide (LiTFSI) since it exhibits high electrochemical stability, indicating its potential as a suitable electrolyte for safer Li ions cells without compromising the performance [24]. $Li_4Ti_5O_{12}$ (LTO) anode and $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ (NMC) cathode batteries exhibited a capacity of 165 mAh·g^{-1} at a rate of 0.1C, and more than 98% capacity was retained after 200 cycles at a rate of 0.5C [25]. ADN also provides excellent temperature stability from $-30\ °C$ to $180\ °C$.

2.4. Fire-Extinguishing Electrolyte

A new trend in the search for safer batteries is to design self-cooling and flame-retarding electrolytes for the battery. According to Jiang et al., "cooling is the key to curbing thermal runaway and compatibility is the basis to ensure electrochemical performance" [9]. They designed a self-cooling composite electrolyte which possessed flame-retardant properties. The thermal stability of the proposed electrolyte was improved by adding *N,N*-dimethylacetamide (DMAC) as a Lewis base and perfluoro-2-methyl-3-pentanone (PFMP) as a self-cooling micro-fire-extinguisher. Fluorocarbon surfactant (FS) was introduced to improve the interface compatibility and electrochemical performance of the battery. The improved thermal stability and electrochemical performance of the Li/C and NMC/Li half cells and full cells were observed with the developed electrolyte. Shi et al. synthesized the electrolyte by mixing propylene carbonate with 1,1,2,2-tetrafluoroethyl-2,2,3,3-tetrafluoropropyl ether at 25 °C with a conductivity of 5.04 mS·cm^{-1} [16]. This electrolyte exhibited good safety features and wettability to electrodes and separator. The batteries also exhibited decent cycle performance at high (60 °C) and low temperature (-40 °C). They also synthesized a non-flammable hydrofluoroether electrolyte, 1,1,2,2-tetrafluoroethyl-2,2,3,3-tetrafluoropropyl ether (F-EPE), by mixing lithium bis(oxalato)borate (LiBOB) and gamma-butyrolactone(GBL) for Li-ion batteries [10]. The addition of F-EPE to the electrolyte exhibited an increased level of safety, decreased surface tension, and good wettability. The electrolyte supported the graphite/$LiCo_{1/3}Mn_{1/3}Ni_{1/3}O_2$ full cell in achieving capacity retention of 80.6% after 500 cycles at room temperature. The battery also delivered a high capacity of 74.2 mAh·g^{-1} at 40 °C.

2.5. Fluorinated/Phosphonate Electrolyte

Fluorinated/phosphonate-based electrolytes are discussed in this section. The flammability of two thermally stable and non-flammable all-fluorinated electrolytes that do not burn on ignition was compared by Fan et al. [26]. The electrolyte was made up of 1 M lithium hexafluorophosphate ($LiPF_6$) in fluoroethylene carbonate/3,3,3-fluoroethylmethyl carbonate/1,1,2,2-tetrafluoroethyl-2′,2′,2′-trifluoroethyl ether (FEC:FEMC:HFE, *w/w/w* = 2/6/2). Figure 4 shows the comparison of the flammability tests of 1 M $LiPF_6$ EC/DMC, 1 M $LiPF_6$ FEC/DMC, and 1 M $LiPF_6$ FEC/FEMC/HFE. It was determined that the fluorine substitution on the alkyl moiety inhibited the propagation of oxygen radicals during combustion.
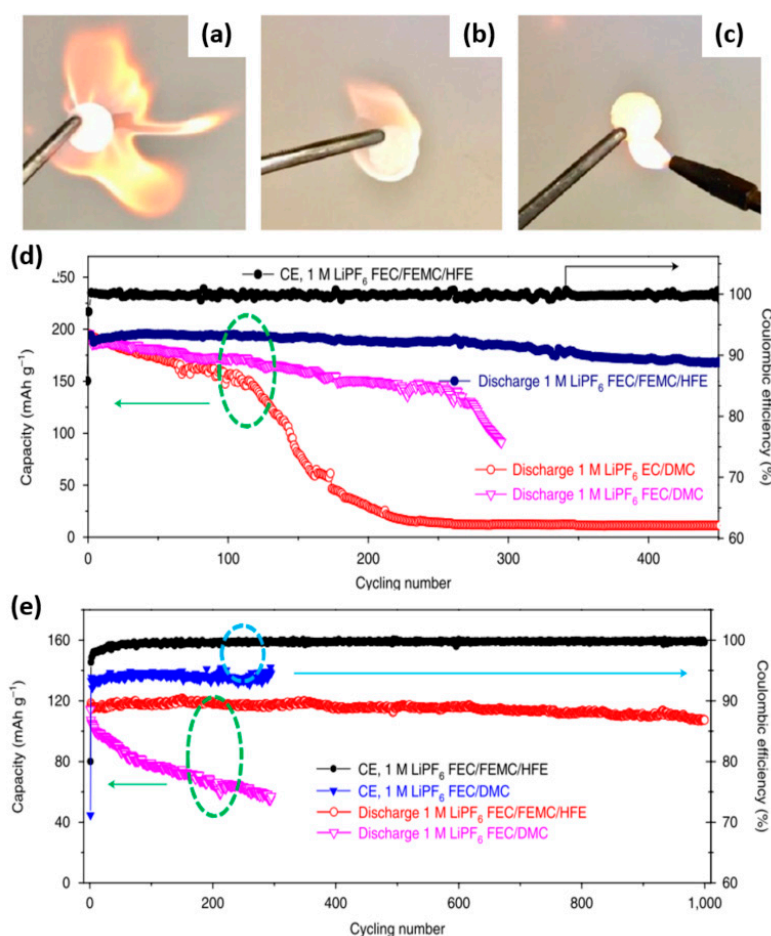
Figure 4. Flammability test for (**a**) 1 M LiPF$_6$/EC/DMC, (**b**) 1 M LiPF$_6$/FEC/DMC, and (**c**) 1 M LiPF$_6$/FEC/FEMC/HFE, (**d**) Cycling stability at 0.5C; (**e**) cycling stability at 1C [26]. Reprinted with permission from Springer Nature, Nature; X. Fan, L. Chen, O. Borodin, X. Ji, J. Chen, S. Hou, T. Deng, J. Zheng, C. Yang, S. Liou, K. Amine, K. Xu, and C. Wang, Batteries with aggressive cathode chemistries, Nat. Nanotechnology, © 2018.

The study claimed that the LiPF$_6$ FEC/FEMC/HFE (fluoroethylene carbonate/3,3,3-fluoroethylmethyl carbonate/1,1,2,2-tetrafluoroethyl-2′,2′,2′-trifluoroethyl ether) electrolyte supported high-voltage cathodes of Li-metal batteries and improved Li plating/stripping, reduced dendrite formation, and increased the battery safety. Their full cells retained 93% capacity even after 1000 cycles at high voltage and a loading of 2 mAh·cm^{-2}, which was credited to the formation of the fluorinated interface. In a similar study, Zeng and coworkers adjusted the molar ratio of Li salt to the solvent to ~(1:2) to obtain an improved non-flammable phosphate electrolyte with better compatibility with the graphite anode [27]. In total, 18,650 Li-ion cells with this electrolyte exhibited a safe and good cycle life and high cyclability with a Coulombic efficiency of 99.7%. The electrolyte was less reactive to Li-metal electrodes; hence, good stability and a high Coulombic efficiency of >99% was obtained for Li-metal plating and stripping in Li–Cu half cells. The safety performance of the batteries was tested by performing short-circuiting, crushing, and nail penetration tests. Figure 5 shows the comparison between the cells using the high-molar-ratio electrolyte and the commercial carbonate electrolyte. The 1:2 ratio phosphate electrolyte passed all three tests, whereas the commercial electrolyte passed only the crush test. They also compared the flame tests of different electrolytes and, as shown in the figure, 1:2 LiTFSI + FEC + LiBOB was the most non-flammable.
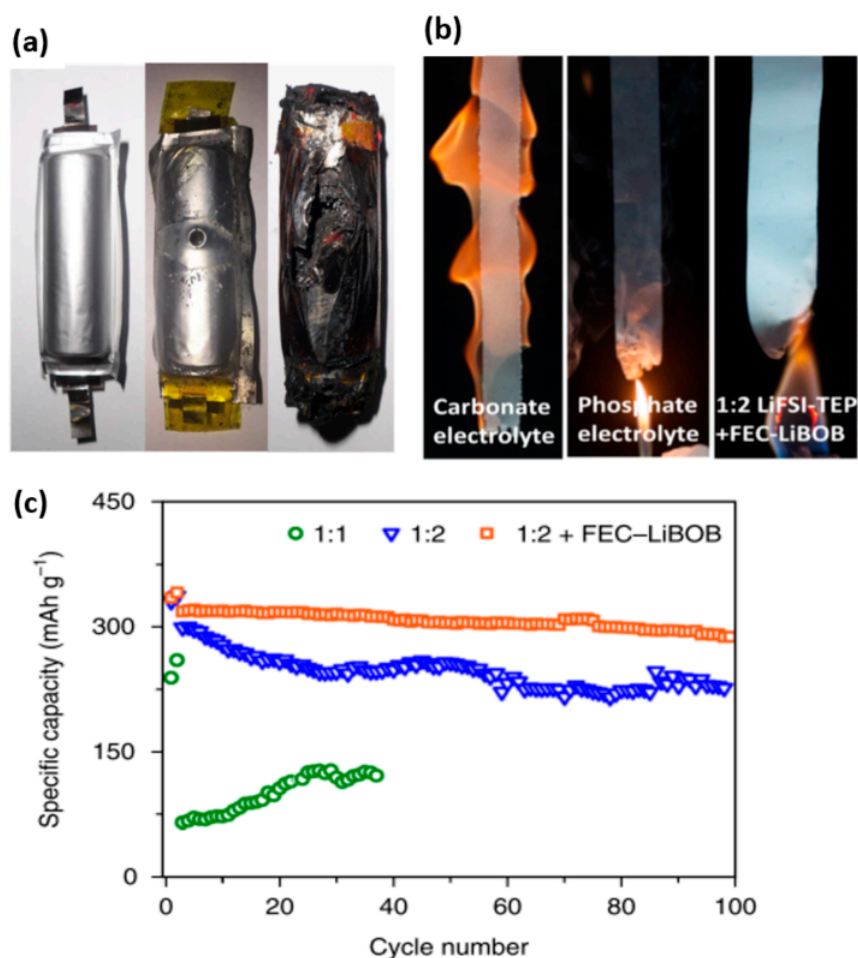
Figure 5. (**a**) Blank cell before nail penetration test for 18,650 cells using 1:2 LiFSI–TEP (Triethyl phosphate) + FEC–LiBOB electrolyte (middle) and commercial carbonate electrolyte (1.0 M $LiPF_6$/EC:DEC:EMC, $v/v/v$ = 1/1/1). (**b**) Flame tests of 1.0 M $LiPF_6$/EC:DEC:EMC ($v/v/v$ = 1/1/1) electrolyte, 1:2 LiFSI–TEP electrolyte, and 1:2 LiFSI–TEP + FEC-LiBOB electrolyte. (**c**) Cycling performance at current density of 20 mA·g^{-1} with different electrolytes [27]. Reprinted with permission from Springer Nature, Nature Energy; Z. Zeng, V. Murugesan, K.S. Han, X. Jiang, Y. Cao, and L. Xiao, Non-flammable electrolytes with high salt-to-solvent ratios for Li-ion and Li-metal batteries; © 2018.

To overcome the risk of battery fires and explosions, Pham et al. developed a novel non-flammable electrolyte composed of 1 M Li-hexafluorophosphate salt, propylene carbonate, and fluorinated linear carbonate co-solvents with high voltage stability [28]. The full batteries with the electrolyte with 1 wt.% FEC additive displayed high energy density, and stable cyclability performance at high voltages up to 5 V. Figure 6 shows the comparison between the fire tests of various compositions of electrolytes. The study reported that the addition of di-(2,2,2 trifluoroethyl) carbonate (DFDEC) enabled the cathode to charge to 5.0 V and to deliver a significantly increased capacity of 250 mAh·g^{-1} or higher.
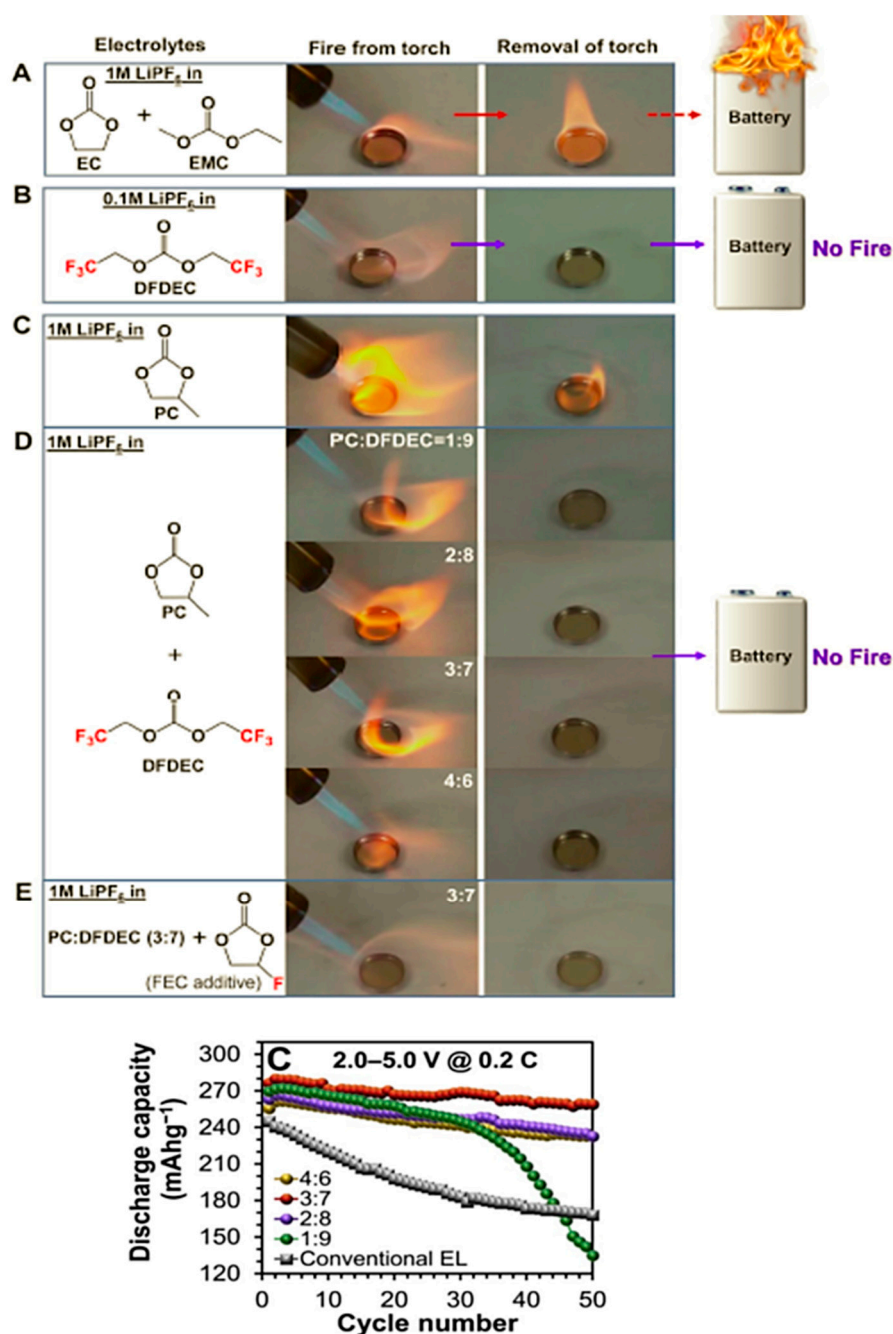
Figure 6. Fire test results for (**A**) conventional electrolyte of 1 M LiPF$_6$/EC:EMC (3:7 vol ratio), (**B**) 0.1 M LiPF$_6$/di-(2,2,2 trifluoroethyl) carbonate (DFDEC), (**C**) 1 M LiPF$_6$/propylene carbonate (PC), (**D**) designed electrolytes of 1 M LiPF$_6$/PC:DFDEC at volume ratios of 1:9, 2:8, 3:7, and 4:6, and (**E**) 1 M LiPF$_6$/PC:DFDEC (3:7) with 1 wt.% FEC additive. The charge–discharge cycling performance is shown below [28]. Reprinted with permission by Elsevier from Non-flammable organic liquid electrolyte for high-safety and high-energy density Li-ion batteries; H. Quang, H. Lee, E. Hwang, Y. Kwon, and S. Song, J. Power Sources, © 2018.

In recent years, the development of non-flammable electrolytes using trimethyl phosphate (TMP) was a topic of interest for research due to its good oxidation and poor reduction stability at the graphite anode. An electrolyte with vinylene carbonate, vinyl ethylene carbonate, and used cyclo-hexane as additives improved the battery performance [29]. A mixed electrolyte with 2 M LiPF$_6$ in TMP was developed for a Li-ion battery with capacity retention of 97% at 50 cycles [30]. Li salt concentration

was optimized, which resulted in improved battery safety. A novel non-flammable electrolyte of $LiPF_6$ with dimethyl methyl-phosphonate (DMMP) and lithium bis-oxalatoborate (LiBOB) was reported by Dalavi's group [31]. Dimethyl methyl-phosphonate was used as a flame retardant and lithium bis-oxalatoborate was used as an anode-forming additive. The developed electrolyte exhibited improved thermal stability as compared to $LiPF_6$ in EC/EMC without affecting the conductivity. The battery cycling performance was also significantly improved. Fluorinated alkyl phosphates (FAP) were used for safer electrolytes [32]. $LiPF_6$ with tris(2,2,2-trifluoroethyl) phosphate (TFEP) solvent was combined with alkyl carbonates such as EC and fluoroethylene carbonate (4-fluoro-2-oxo-1,3-dioxolane, FEC). Addition of EC as a co-solvent exhibited high exothermic response at onset temperatures below 200 °C, whereas FEC showed lower exothermic peaks at higher onset temperatures above 250 °C; hence, they concluded that FEC is a better co-solvent for safer Li-ion batteries for severe conditions.

Non-flammable electrolyte was obtained by Kurc via the dissolution of solid lithium bis(trifluoromethane sulphonyl) imide ($LiNTF_2$), lithium bis-oxalato borate (LiBOB), and lithium hexafluorophosphate ($LiPF_6$) in tetrahydrothiophene 1,1-dioxide or tetramethylene sulfone (TMS) with 10% vinylene carbonate (VC) [33]. They used $LiNiO_2$ as a cathode and the 1 M $LiPF_6$ in TMS + 10% VC electrolyte for the Li-ion battery and obtained 195 cycles and a Coulombic efficiency of 140 mAh·g^{-1} after 20 cycles at a rate of C/10. TMS has good chemical and thermal stability, a low autoprotolysis constant, high polarity, low toxicity, low vapor pressure, and a high melting point of 275 °C. To overcome the challenge of obtaining a poly (ethylene oxide) (PEO)-based solid electrolyte with high Li-ion conductivity and good mechanical strength, Li et al. synthesized a polymeric electrolyte with 40% poly-methyl hydrogen-siloxane (PMHS) and PEO [34]. The membrane exhibited an ionic conductivity of 2.0×10^{-2} S·cm^{-1} at 80 °C, a high electrochemical window, and better thermal stability. The assembled Li/$LiFePO_4$ battery exhibited good stability and obtained a reversible capacity of 140 mAh·g^{-1} at 60 °C at a rate of 0.1C. A non-flammable phosphate-based electrolyte composed of 5 M L1 (M) Li bis(fluorosulfonyl) imide (LiFSI) in a trimethyl phosphate (TMP) solvent was developed Shi et al. [35]. Figure 7 depicts that the electrolyte with 1 M LiPF6/ethylene carbonate (EC) + diethyl carbonate (DEC) ignited immediately and continued to burn, whereas the 5 M LiFSI/TMP electrolyte was non-flammable. Four hundred cycles were obtained when the electrolyte was used in Li | $LiFePO_4$ cells.
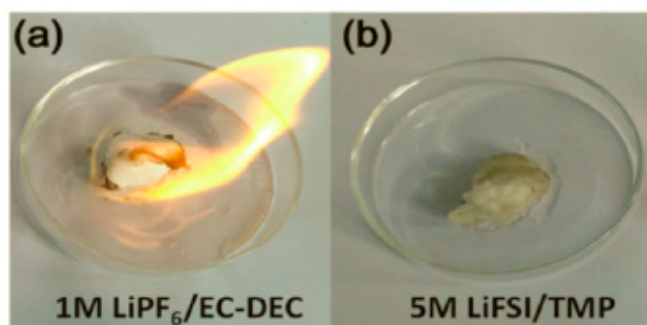

Figure 7. (**a**) Flammability tests of 1 M $LiPF_6$/EC-DEC and (**b**) 5 M LiTFSi/TMP electrolyte [35]. Reprinted with permission, © 2018.

Janssen et al. synthesized 1,3-dimethylimidazolidin-2-mm-trifluoroborate ($NHC-BF_3$) and 1,3-dimethylimidazolidin-2-mm-tetrafluorotrifluoromethylphosphate ($NHC-PF_4CF_3$) on $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ (NMC111) electrodes of Li-ion batteries as overcharge protection shutdown additives in 1 M $LiPF_6$ in EC:DEC (w/w = 3/7) electrolyte [36].

2.6. Gel Polymer Electrolytes

Gel polymer electrolytes (GPE) are made by combining different types of salts such as $LiPF_6$ and $LiBF_4$ in aprotic solvents like PC, EC:DEC, and EC:DMC, prepared by ultraviolet (UV) polymerization. They have characteristics of both solid and liquid electrolytes. Various GPEs with different compositions were developed for different battery chemistries [37–40]. In this section, we discuss the polymer electrolytes that were fabricated for safer Li-ion batteries. Free-standing gel polymer electrolytes (GPE) with polymeric ionic liquid, poly[diallyldimethylammonium] bis(trifluoromethane) sulfonimide, 1-ethyl-3-methylimidazolium bis (trifluoromethane) sulfonimide, and lithium salt were developed by Meer et al. [15], and were found to be thermally stable. The electrolyte contained 80 wt.% ionic liquid and lithium salt content, which resulted in highly ionically conductive electrolyte films. Galvanostatic charge/discharge cycling of Li/GPE/LiFePO$_4$ batteries exhibited a discharge capacity of 169.3 mAh·g^{-1} at C/10 and 126.8 mAh·g^{-1} at 1C. The capacity retention capability at a rate of 5C was up to 40 cycles at 22 °C. Figure 8 shows the free-standing GPE.
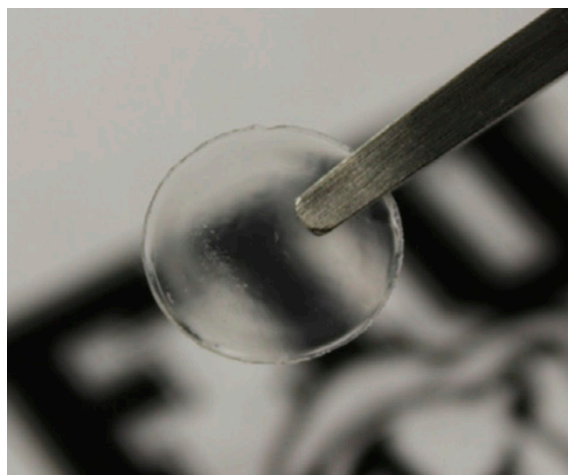

Figure 8. Gel polymer electrolyte (GPE) demonstrating its free-standing property [15]. Reprinted with permission by Elsevier from Electrochimica Acta, 213, Meer Safa, Amir Chamaani, Neha Chawla, and Bilal El-Zahab, Polymeric Ionic Liquid Gel Electrolyte for Room Temperature Lithium Battery Applications, 587–593, © 2016.

A gel polymer electrolyte (GPE) based on electro-spun polyvinylidene fluoride (PVDF)/halloysite nanotube (HNT) nanocomposite non-wovens was synthesized and used as a separator for Li-ion batteries [41]. The GPE as a separator exhibited minimal thermal shrinkage and higher melting temperature which was proven to be important for obtaining a safer Li-ion battery. A good tensile and puncture strength as compared with a commercially available separator was observed. The developed Li/GPE/LiCoO$_2$ battery obtained a capacity of 138.01 mAh·g^{-1} with 97% Coulombic efficiency. The batteries even displayed better cyclability.

A nano-particle-decorated poly(methyl methacrylate-acrylonitrile-ethyl acrylate) (P(MMA-AN-EA))-based gel polymer electrolyte (GPE) was formulated using 1-ethyl-3-methylimidazolium bis(trifluoromethanesolfonyl) imide (EMITFSI) ionic liquid as a plasticizer by Li et al. [13]. The membrane exhibited a high porosity of 70%, an ionic conductivity of 3.2×10^{-3} S·cm^{-1} at ambient temperature with better anti-thermal shrinkage and flame retardation performance, and a fracture strength of 160 MPa. The batteries fabricated with the membrane retained ~95% capacity after 100 cycles under 0.2C at room temperature. Figure 9 shows the flame test of the membrane soaked with commercially carbonated organic electrolyte and the ionic liquid.
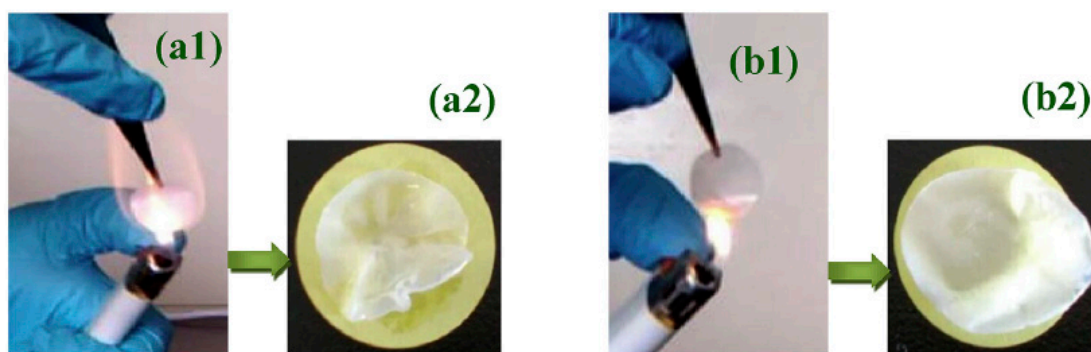
Figure 9. Combustion test of the MS5 (SiO_2:Al_2O_3 = 5:5) membrane immersed in commercial carbonated organic liquid electrolyte before (**a1**), and after (**a2**) the burning test; and the combustion test of the membrane immersed in the ionic liquid electrolyte before (**b1**), and after (**b2**) the burning test [13]. Reprinted with permission by Elsevier from Application of the imidazolium ionic liquid based nano-particle decorated gel polymer electrolyte for high safety lithium ion battery; M. Li, Y. Liao, Q. Liu, J. Xu, P. Sun, and H. Shi; Electrochimica Acta, 284 188–201, © 2018.

A gel polymer electrolyte containing succinonitrile (GPE-SN) was designed using the immersion method by Pengfei Lv, represented as GPE-SN-IM. GPE-SN-IM exhibited an ionic conductivity of 1.63×10^{-3} S·cm^{-1} at 25 °C [42]. It obtained a tensile strength of 6.5 MPa. A $LiCoO_2$/$Li_4Ti_5O_{12}$ film battery with a high-mechanical-strength GPE-SN-IM film displayed a good cyclability of 100 cycles. These batteries showed good cyclability even at elevated temperatures up to 55 °C due to better thermal stability. Figure 10 shows the flammability tests of GPE-LE with EC/DMC and GPE-SN-IM, proving that GPE-SN-IM is a much safer electrolyte for Li-ion batteries.
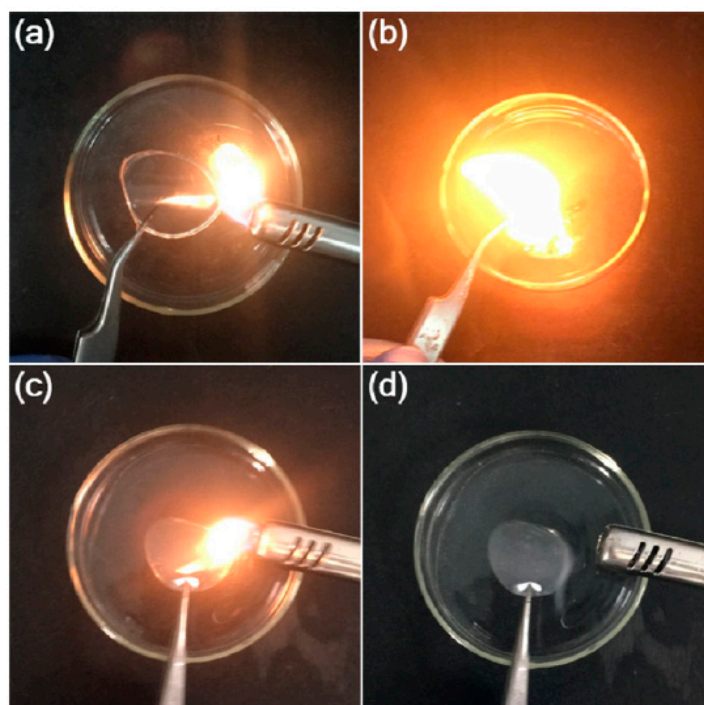

Figure 10. Flame ignition tests for (**a**) GPE-LE and (**c**) GPE-SN-IM. After contacting the flame, (**b**) GPE-LE quickly burns, but (**d**) GPE-SNIM does not burn [42]. Reprinted with permission by the American Chemical Society from Robust Succinonitrile-Based Gel Polymer Electrolyte for Lithium-Ion Batteries Withstanding Mechanical Folding and High Temperature; P. Lv, Y. Li, Y. Wu, G. Liu, H. Liu, S. Li, C. Tang, J. Mei, and Y. Li; © 2018.

Polyacrylonitrile (PAN)/poly(vinyl alcohol) (PVA) blending membrane-based gel polymer electrolytes with PVA/PAN ratios of 0:100, 10:90, 20:80, and 40:60 were developed, denoted as BM-0, BM-1, BM-2, and BM-3, respectively [43]. The cell test revealed capacity retentions of 58%, 1%, 94%, 96%, and 40% for the Celgard 2320 separator, BM-0, BM-1, BM-2, and BM-3, respectively, after 200 cycles at a rate of 1C. Figure 11 shows the images of the membranes heated from 25 °C to 100 °C to 160 °C. Celgard 2320 shrinked severely at 160 °C, while BM-0, BM-1, BM-2, and BM-3 showed a negligible dimensional change due to the PAN membrane, which possesses a melting temperature of more than 300 °C.
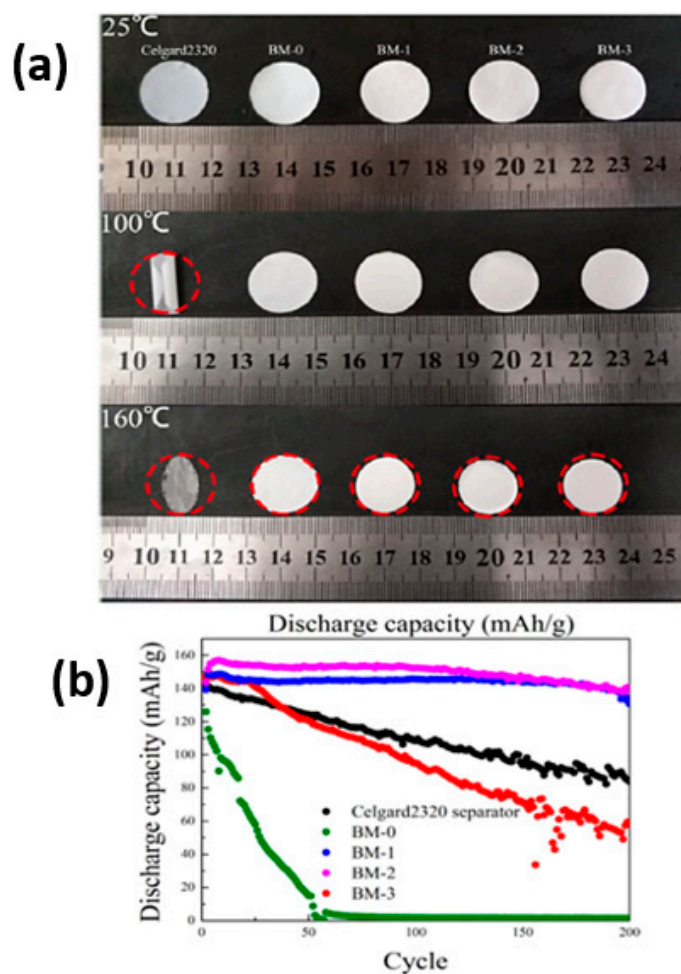

Figure 11. (**a**) Photographs of the membranes before and after heat treatment in an oven at 25 °C, 100 °C, and 160 °C. (**b**) The cycling stability of the cells at 1C [43]. Reprinted with permission by Elsevier from Blending-based polyacrylonitrile/poly (vinyl alcohol) membrane for rechargeable lithium ion batteries; C. He, J. Liu, J. Li, F. Zhu, and H. Zhao, J. Memb. Sci.; 560; © 2018.

In situ thermally induced free-radical polymerization was used to develop a GPE (gel polymer electrolyte) with an interpenetrating polymer network (IPN-GPE) [44]. The IPN-GPE with cross-linked structure exhibited high flexibility and deformability, and a thermal stability of over 310 °C. It displayed a good electrochemical stability window and an ionic conductivity of 1.3×10^{-3} S·cm^{-1} at 30 °C. The rechargeable batteries with the gel electrolyte displayed good electrical performance, safety, cyclability, and rate capability. Combustion tests were performed on conventional carbonate-based, 1 M LiPF$_6$ in EC/DMC/DEC ($v/v/v$ = 1/1/1) liquid electrolyte and IPN-GPE, as depicted in Figure 12. The low flammability of IPN-GPE makes it a safer electrolyte for Li-ion batteries.
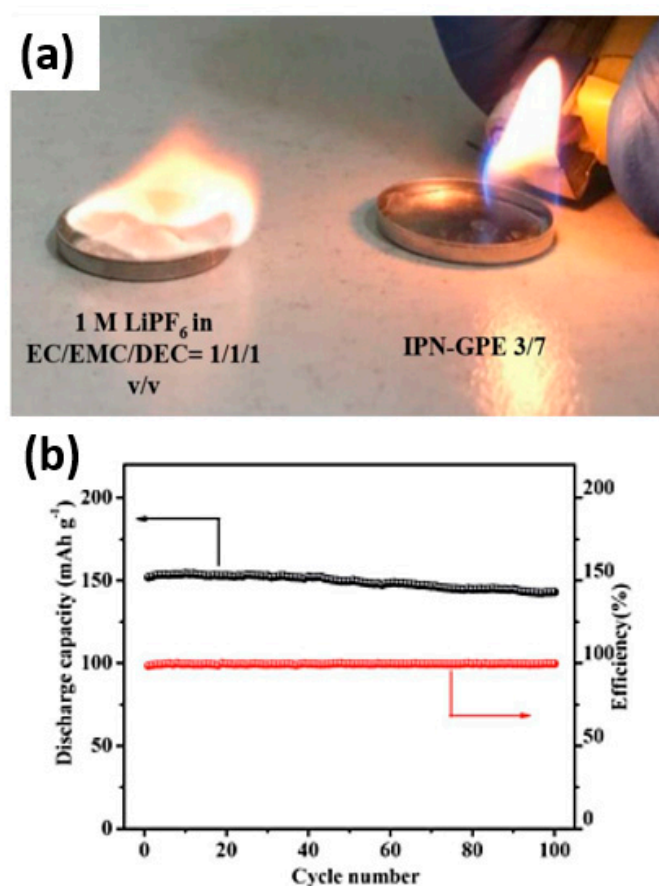
Figure 12. (**a**) Combustion testing of 1 M LiPF$_6$ EC/EMC/DEC liquid electrolyte and IPN-GPE (Gel polymer electrolyte with interpenetrating polymer network). (**b**) The cycling performance of the cell at 0.1C [44]. Reproduced from Reference [44] with permission from the Royal Society of Chemistry.

The effect of adding fluoroethylene carbonate (FEC) as an electrolyte additive for ethyl-methyl sulfone (EMS) electrolytes with LiPF$_6$ as a conducting salt in graphite based batteries was studied, and it was concluded that lowering the EC led to a lower conductivity of the electrolyte, whereas increasing EC enhanced flammability [45]. Wang et al. developed and studied a star-comb co-polymer based on poly(D,L-lactide) (PDLLA) macromonomer and poly(ethylene glycol)methyl ether methacrylate (PEGMA) for Li-ion batteries [46]. For synthesis of the six-arm vinyl-functionalized PDLLA macromonomer, a ring-opening polymerization (ROP) of D,L-lactide and an acylation of the hydroxy end-groups were used. The prepared solid polymer electrolyte exhibited good thermal stability and electrochemical properties. A gel electrolyte consisting of a fluoropolymer/cellulose derivative matrix and liquid electrolyte was fabricated with reversible thixotropic transformation and abuse tolerance [47]. The electrolyte displayed high ionic conductivity and low crystallinity. Flexible composite ionic liquid gel polymer electrolytes (ILGPEs) supported by Li$_{1.5}$Al$_{0.5}$Ge$_{1.5}$(PO$_4$)$_3$ (LAGP) were fabricated and investigated by Guo et al. for improving the electrochemical performance and thermal safety of the batteries [48]. They optimized 10% LAGP for the electrolyte. LAGP particles reduced the crystallinity of the polymer matrix, thus providing lithium ions, which resulted in elevated ionic conductivity and Li-ion transference number. The cycling stability of LiFePO$_4$/10% LAGP/Li batteries was improved since no dendrite was formed on the lithium anode. Figure 13 shows the flammability tests and the cycling performance of the batteries.
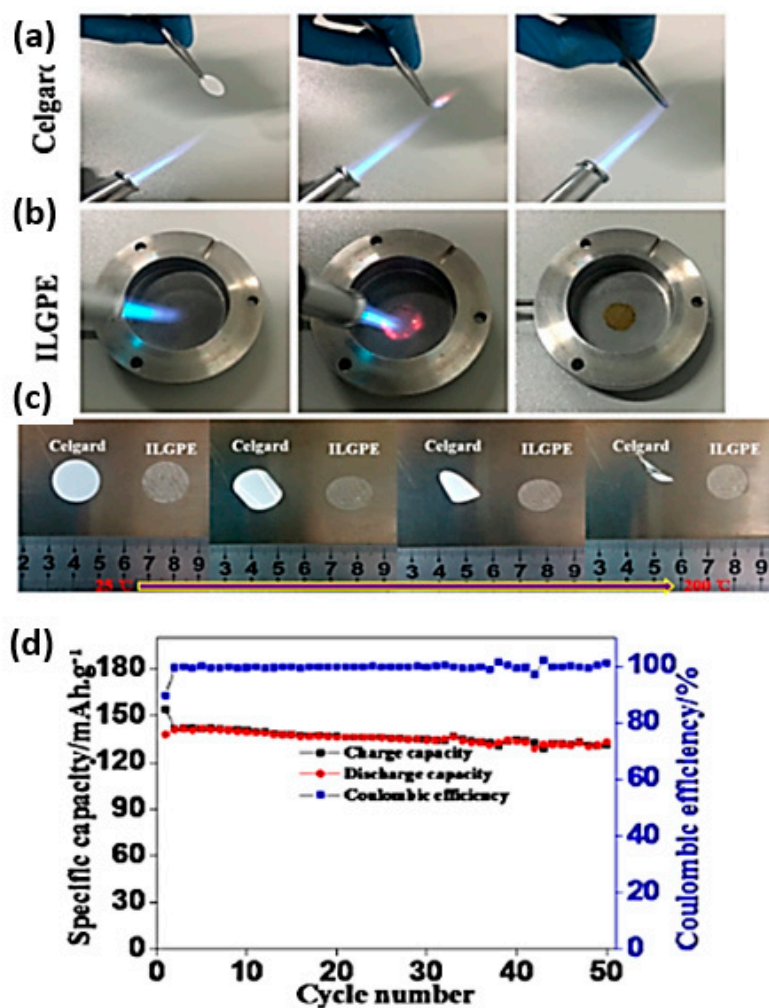
Figure 13. (**a**–**c**): Flammability tests of commercial Celgard membrane and ILGPE/10%LAGP (ionic liquid gel polymer electrolytes /10% $Li_{1.5}Al_{0.5}Ge_{1.5}(PO_4)_3$). (**d**) The cycling performance of $LiFePO_4$/ILGPE/10%LAGP/Li. Reproduced from Reference [48] with permission, © 2018.

Fu et al. developed a bendable, flexible polymer electrolyte membrane (PEM) based on cross-linkable polyurethane precursor, polyethylene glycolbiscarbamate dimethacrylate (PEGBCDMA), that was thermally stable and flame retardant [49]. The lithium iron phosphate ($LiFePO_4$)/PEM/graphite full battery with the PEM exhibited approximately 80% specific capacity retention up to 250 cycles. A cross-linking polymer network of poly(acrylic anhydride-2-methyl-acrylic acid-2-oxirane-ethyl ester-methyl methacrylate) (PAMM)-based electrolyte was developed as an electrolyte material for lithium-ion batteries by Ma et al. [50]. The $LiNi_{0.5}Mn_{1.5}O_4$/Li and $LiNi_{0.5}Mn_{1.5}O_4$/$Li_4Ti_5O_{12}$ batteries with PAMM-based gel polymer electrolyte delivered stable charging/discharging profiles and excellent rate performance at room temperature and even at 55 °C. A capacity of 128 mAh·g^{-1} and a significantly improved Coulombic efficiency of 96% at 0.1C after 100 cycles for $LiNi_{0.5}Mn_{1.5}O_4$/$Li_4Ti_5O_{12}$ was achieved, as shown in Figure 14.
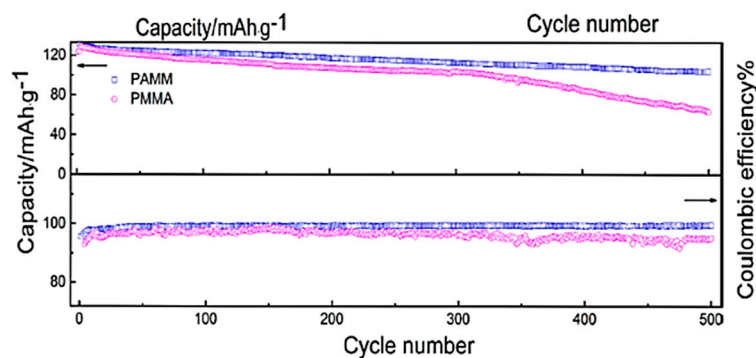
Figure 14. Discharge capacity and Coulombic efficiency of LiNi$_{0.5}$Mn$_{1.5}$O$_4$/Li cells using PAMM (poly(acrylic anhydride-2-methyl-acrylic acid-2-oxirane-ethyl ester-methyl methacrylate)) and PMMA (poly(methyl methacrylate-acrylonitrile) at 0.1C [50]

A flexible, flame-retardant solid polymer electrolyte, combining PCL/SN (Poly(ε-caprolactone)/ Succinonitrile) blends with a polyacrylonitrile (PAN)-skeleton and forming hierarchical architectures, was developed by Zhang et al. [51]. Kim et al. used Mg(OH)$_2$, a flame-retardant material, for their composite polymer electrolyte (CPE) [52]. They did a comparative study by varying the amount of Mg(OH)$_2$ added to the electrolyte. Figure 15 shows the flammability tests at different wt.% additive in the electrolyte. As can also be seen from Figure 15, the composite gel polymer electrolyte with 40 wt.% Mg(OH)$_2$ displayed a higher discharge capacity at 2C/2C charging/discharging current after 200 cycles, as compared to 20 wt.% or 0 wt.%.
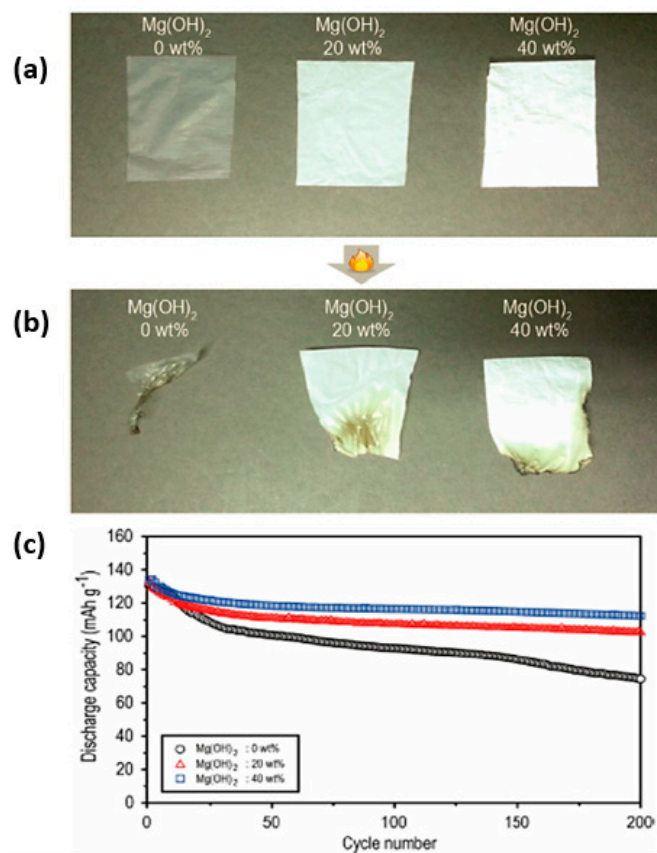

Figure 15. (**a**,**b**) Flame-retardant tests of composite gel polymer films with different amounts of Mg(OH)$_2$. (**c**) The cycle performance of cells with composite gel polymer electrolytes with different concentrations of Mg(OH)$_2$ at 2C rate [52]. Reprinted with permission, © 2018.

An ionogel electrolyte was fabricated by Chen et al. by immobilizing ionic liquids within a nano-porous zirconia-supported matrix (represented as ZIE) which provided the properties of both solid and liquid electrolyte ionic transport, thus improving the thermal stability and safety [53]. The assemble battery exhibited excellent cyclability and a discharge capacity of ~136 mAh·g^{-1} after 200 cycles at 30 °C. The battery had a wide operating temperature range from −10 °C to 90 °C. Figure 16 shows the burning test performed on the ZIE to demonstrate the safety feature. It confirmed the high heat-resistant ability of the electrolyte.
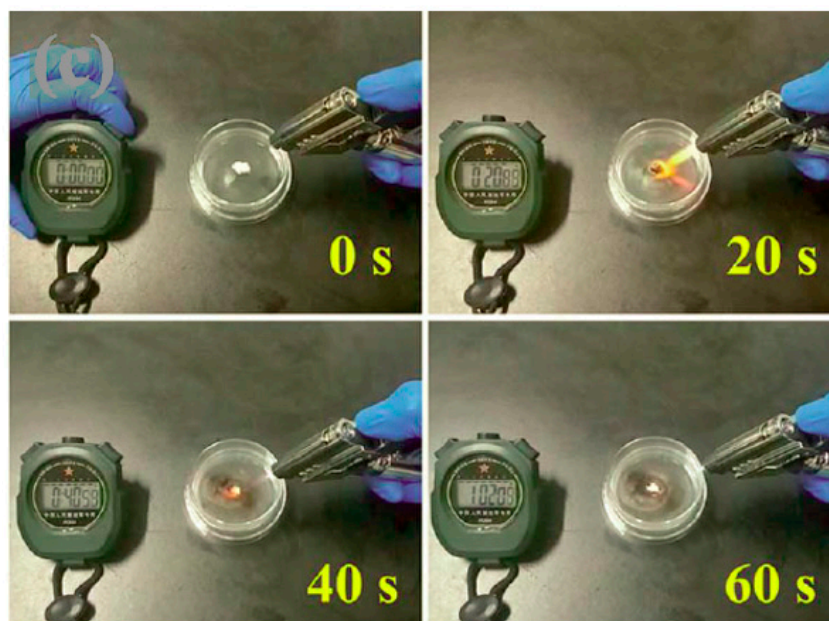

Figure 16. Flammability test of the ZIE using a flame gun [53]. Reproduced from Reference [35] with permission from the Royal Society of Chemistry.

Tetra PEG gel mixed with 1.0 M LiPF$_6$ in an EC + DEC + TFEP mixture ($v/v/v$ = 53/27/20) electrolyte for Li-ion batteries was developed by Han et al. [54]. The resulting Tetra PEG gel electrolyte was used with a LiFePO$_4$ cathode in Li-ion batteries. Li et al. developed a Li-ion battery using an interpenetrating rigid flexible poly (aryl ether ketone) non-wovens (PAEKNW) cross-linked with a poly(ethylene glycol) dimethacrylate electrolyte, LiFePO$_4$ cathode, and Li metal anode [55]. The cross-linked electrolyte exhibited good flame-retarding abilities. Jia et al. chemically bonded phosphonate with the gel polymer electrolyte to obtain a flame-resistive electrolyte for a safe Li-ion battery [56].

2.7. Additives for Electrolytes

Using additives in electrolytes is a proven method to enhance the performance of the battery. Liu et al. used different concentrations of methyl diethyl phospho-noacetate (MDPCT), triethyl-2-fluoro-2-phosphonoacetate (TFPCT) and carbethoxy ethylidene triphenylphosphorane (CETPE) flame-retardant additives into the blank electrolytes to improve the thermal stability of the electrolyte and reduce the flammability [57]. They found 5 wt.% additive to be the optimum for the most flame-retardant effect. According to the electrochemical tests, the addition of 5 wt.% TFPCT enhanced the discharge capacity of the NCA (LiNi$_{0.8}$Co$_{0.15}$Al$_{0.05}$O$_2$)/Li half-cell, which exhibited a capacity retention as high as 92.2% at a current density of 0.5C after 100 cycles. Figure 17 shows the flammability tests of the various electrolytes.

Figure 17. Flammability measurement of PVDF-HFP (polyvinylidenefluoride-hex- afluoropropylene), PBO (poly [benzyl methacrylate-co-oligo(ethylene glycol)ether methacrylate]), and c-PPO (cross-linked poly[dimethyl-p-vinyl benzyl phosphonate-co-oligo (ethylene glycol) meth acrylate] co-polymer) polymer matrices and their corresponding GPEs.

The addition of 0.5% of diethyl(thiophen-2-ylmethyl) phosphonate (DTYP) additive to the base electrolyte aided in improving the capacity retention of a high-voltage Li-ion cell using $LiNi_{0.5}Mn_{1.5}O_4$ from 18% to 85% after 280 cycles at 1C at 60 °C. The novel electrolyte also aided in reducing the self-extinguishing time of the electrolyte from 88 s to 77 s [58]. Figure 18 shows the flammability tests and the cycling performance of the batteries with and without DTYP electrolyte.
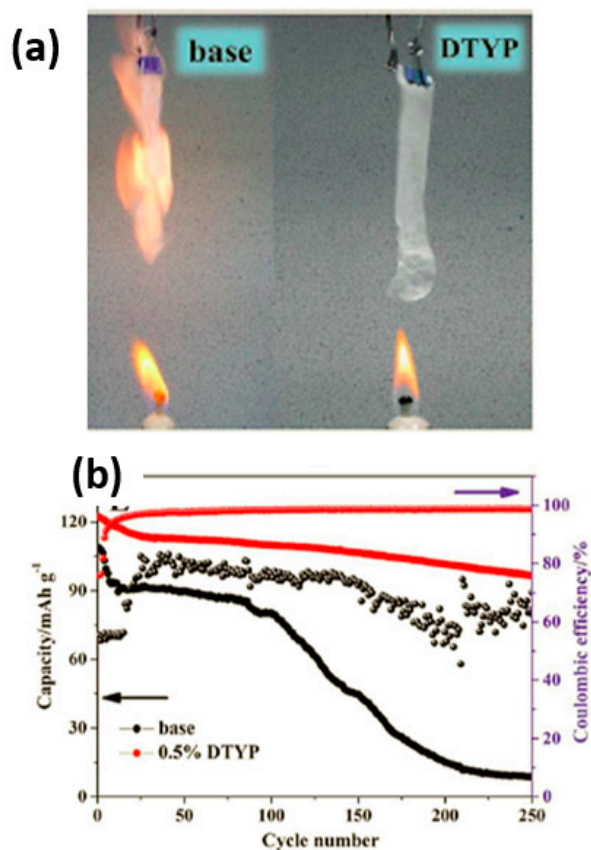

Figure 18. (**a**) Flammability tests of the PE membrane with base and DTYP-containing electrolytes. (**b**) The cycling performance of the batteries with the base and DTYP-containing electrolytes [58]. Reprinted with permission, © 2018.

Sheng et al. improved the ionic conductivity of solid-state electrolytes via an interaction between $Mg_2B_2O_5$ nanowires and SO_2^- in the $TFSi^-$ anion [59]. The $Mg_2B_2O_5$ nanowire additive has high strength and is a good flame retardant. The cycling performance of the solid-state Li-ion batteries with poly (ethylene oxide)/$LiTFSi$/$Mg_2B_2O_5$ electrolyte at 1.0C and 50 °C is shown in Figure 19. A stable specific capacity of approximately 120 mAh·g^{-1} in 230 discharge–charge cycles was achieved.
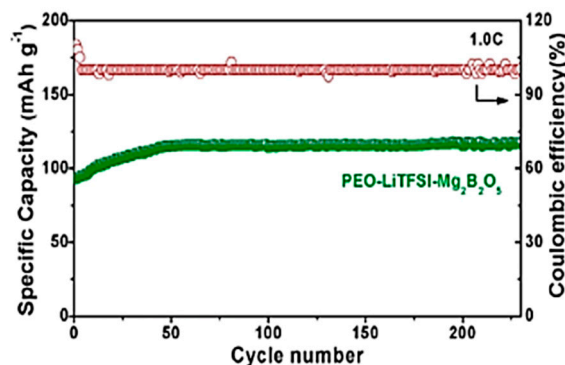
Figure 19. Cycling performance of $LiFePO_4$/Li solid-state Li-ion batteries with poly(ethylene oxide)/$LiTFSi$/10% $Mg_2B_2O_5$ at 50 °C [59], © 2018.

An electrolyte additive of a fluorinated phosphazene derivative, ethoxy-(pentafluoro)-cyclotriphosphazene (PFN), that significantly improved the battery performance of lithium nickel manganese oxide ($LiNi_{0.5}Mn_{1.5}O_4$) cathode batteries was fabricated by Liu et al. [60]. Figure 20 shows the flammability tests comparing the PFN electrolyte with $LiPF_6$ and the cycling performance of a $LiNi_{0.5}Mn_{1.5}O_4$/graphite full cell with or without 5 wt.% PFN-containing electrolyte at 1C.
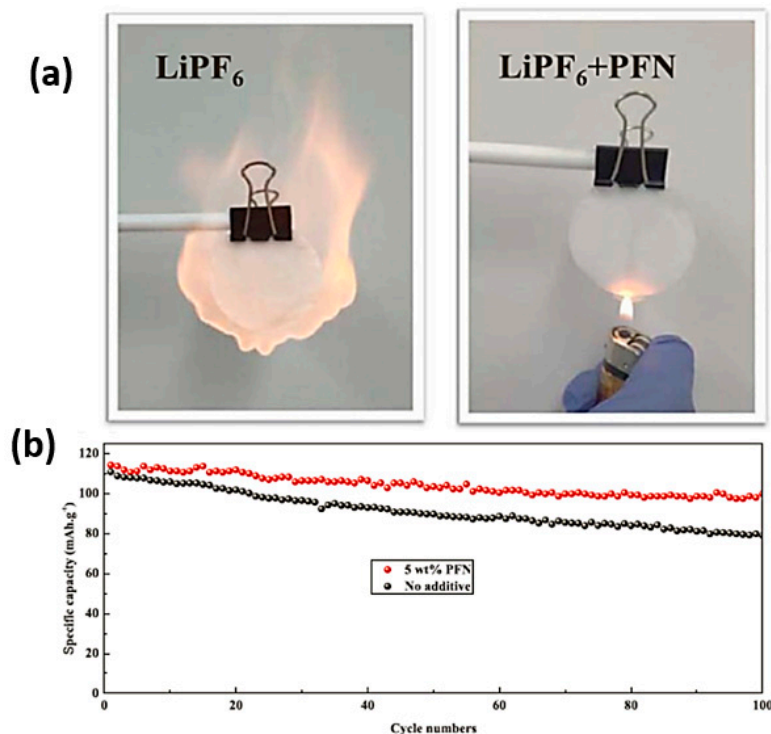
Figure 20. (**a**) Flammability tests of base electrolyte 1 M $LiPF_6$/EC + DEC + DMC (*v/v/v* = 1/1/1) and (**b**) the same electrolyte with 5 wt.% PFN (Fluorinated phosphazene derivative, ethoxy-(pentafluoro)-cyclotriphosphazene) additive. (**c**) The cycling performance of the batteries with and without the electrolyte, and with additive is shown below [60]. Reprinted with permission, © 2018.

A new flame-retardant electrolyte additive ethoxy(pentafluoro)cyclotriphosphazene (PFPN) was developed by Li et al., with 5% additive to the electrolyte achieving better flame-retardant properties [61]. The initial discharge capacity of the Li-ion batteries with $LiCoO_2$ cathode and 5% PFPN was 150.7 mAh·g^{-1}, with a capacity retention of 99.14% after 30 cycles at 0.1C. A poly(bis-(ethoxyethoxyethoxy)phosphazene) (EEEP) electrolyte additive with an electro-oxidable P–O bond was developed by Zhou et al. [62] to enhance the cycling performance of $LiCoO_2$ cathodes under high-voltage operations. The 5 wt.% EEEP in electrolyte aided in reducing the flammability and also increasing the cyclability of the battery by forming a protective layer on the cathode surface that prevented electrolyte decomposition.

There were few researches on water-in-salt electrolytes for replacing the traditional organic electrolytes in Li-ion batteries to improve the safety and stability of the batteries at a lower cost. Sun et al. demonstrated the use of "water-in-salt" electrolyte (21 M LiTFSI in H_2O) with a TiS_2 anode, which displayed high electrochemical reversibility when paired with a $LiMn_2O_4$ cathode [63].

Table 1 summarizes the different non-flammable electrolytes and their electrochemical performance for Li-ion batteries.

Table 1. Summary of non-flammable electrolytes and their electrochemical performance.

Electrolyte Composition	Cathode/Battery Type	Discharge Capacity (mAh·g^{-1})	Number of Cycles	Capacity Retention	Rate	Reference
1 M $LiPF_6$ in EC/DFSM$_2$/EMC (2/3/5 in vol.) + 5 wt% FEC	$LiCoO_2$(LCO)/ graphite full cell		135	92%		[11]
SN1IL/DMC (*v/v* = 1/1) doped with 0.6 M $LiPF_6$/0.4 M LiODFB salts	$LiCoO_2$/graphite full cell	152	~90	72%	2C	[12]
1 M $LiPF_6$ P13-TFSI/EC (1/1) *w/w* 5% FEC	Graphite/Li half batteries	115	~100	95%	C/12	[13]
inorganic non-aqueous liquid electrolyte-$LiAlCl_{4.3}SO_2$ (IE)	LiFePO4	113	~100	94%	5C	[14]
5.3 M LiFSA/TMP	$LiNi_{0.5}Mn_{1.5}O_4$/ graphite/Li half batteries	250	1200	99%	C/5	[23]
ADN + LiTFSI	$LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ (NMC)	165	~200	98%	0.5C	[24]
LiBOB-based electrolyte	graphite/$LiCo_{1/3}Mn_{1/3}Ni_{1/3}O_2$ full cell	108	~500	80%	1C	[10]
1 M $LiPF_6$ in FEC:FEMC:HFE (*w/w/w* = 2/6/2)	Li/LCP cell		1000	93%	1C	[26]
1:2 LiFSI–TEP with FEC–LiBOB composite additives	Li–Cu half cells	135	350	88%	0.05C	[27]
1 M Li-$LiPF_6$ in PC and fluorinated linear carbonate co-solvents	LMNC/graphite full cell	250	100	72%	0.2C	[28]
2 M $LiPF_6$ in EC:DEC:TMP	$LiMn_2O_4$ cathode	34	50	97%	0.2C	[30]
1 M $LiPF_6$ in TMS + 10% VC electrolyte	$LiNiO_2$	145	195			[33]
5 M LiFSI/TMP	Li/LiFePO$_4$ battery	118	400	99%	0.5C	[35]
GPE based on electro-spun PVDF/HNT nano-composite non-wovens	$LiCoO_2$	138	50	97%	0.1C	[41]
P(MMA-AN-EA) + EMITFSI	LiFePO$_4$		100	95%	0.2C	[13]
GPE-SN-IM	$LiCoO_2$/$Li_4Ti_5O_{12}$ film battery	132	100	92%	0.2C	[42]
PAN/PVA (20:80 ratio) blending membrane-based GPE	$LiCoO_2$	160	200	96%	1C	[43]

Table 1. *Cont.*

Electrolyte Composition	Cathode/Battery Type	Discharge Capacity (mAh·g^{-1})	Number of Cycles	Capacity Retention	Rate	Reference
IPN-GPE with 1 M LiPF$_6$ in EC/DMC/DEC $\frac{1}{4}$ 1/1/1, *v/v/v*, liquid electrolyte	LiFePO$_4$	143	100	94%		[44]
4% FEC to the 1 MLiPF$_6$ in EMS electrolyte	NMC/graphite cells	325	100	99%	0.1C	[45]
LFP/PDLLA–SPE/Li	LiFePO$_4$	144.7	250	87%		[46]
1.0 M LiPF$_6$ in EC/DMC mixed with 700 wt.% polymer	LiCoO$_2$/graphite electrodes		300	74%	0.1C	[47]
ILGPEs supported by 10% LAGP	LiFePO$_4$	131	50		0.05C	[48]
PEM based on PEGBCDMA	LiFePO$_4$	125	250	80%	C/3	[49]
PAMM-based gel polymer electrolyte	LiNi$_{0.5}$Mn$_{1.5}$O$_4$/Li and LiNi$_{0.5}$Mn$_{1.5}$O$_4$/ Li$_4$Ti$_5$O$_{12}$ batteries	128	100	96%	0.1C	[50]
Solid polymer electrolyte: PCL/SN blends with PAN-skeleton	LiFePO$_4$	101	400	100%	1C	[51]
40 wt.% Mg(OH)$_2$ added to electrolyte	LiCoO$_2$/graphite	112	200	83%	0.5C	[52]
Ionogel electrolyte	LiFePO$_4$	136	200			[53]
Tetra PEG gel mixed with 1.0 M LiPF$_6$ in an EC + DEC + TFEP mixture (*v/v/v* = 53/27/20) electrolyte	LiFePO$_4$	128	10	95%	0.1C	[54]
PAEKNW) cross-linked with poly(ethylene glycol) dimethacrylate electrolyte	LiFePO$_4$	128	200	90%	1C	[55]
5 wt.% TFPCT	NCA/Li half-cell	120	100	92%	0.5C	[57]
0.5% DTYP additive in base electrolyte	LiNi$_{0.5}$Mn$_{1.5}$O$_4$/ graphite full cells	125	280		1C	[58]
poly (ethylene oxide)-LiTFSi-Mg$_2$B$_2$O$_5$ electrolyte	LiFePO$_4$	120	230		1C	[59]
5 wt.% PFN-containing electrolyte	LiNi$_{0.5}$Mn$_{1.5}$O$_4$ cathode	~110	100		1C	[60]
5% PFPN	LiCoO$_2$	150.7	30	99%	0.1C	[61]
5 wt.% EEEP in electrolyte	LiCoO$_2$		100	91%		[62]

3. Conclusions and Future Direction

Recently, numerous Li-ion battery fire and explosion incidents attracted attention to the issue of battery safety. In this paper, we discussed research conducted toward the development of non-flammable electrolytes to design safer batteries. The use of self-cooling and fire-retardant materials in electrolytes can be an attractive option. The use of fluorinated carbonates and ionic liquids can be potentially useful in designing safer batteries. A fire-extinguishing electrolyte with high cyclability is very interesting for high-performance rechargeable batteries. We hope this article will provide an insight into recent development for safer electrolytes and lithium ion batteries.

Author Contributions: Conceptualization, investigation, original draft preparation, writing review: N.C., editing, proofing & visualization: N.B., S.S., N.C.

Abbreviations

The following abbreviations are used in this manuscript:

Abbreviation	Full form/Chemical Name	Abbreviation	Full form/Chemical Name
LiPF$_6$	lithium hexafluorophosphate	DEC	diethyl carbonate
DMC	dimethyl carbonate	EMC	ethyl methyl carbonate
EC	ethylene carbonate	FEC	fluoroethylene carbonate
MFSM$_2$	fluoro(3-(2-(2-methoxyethoxy)ethoxy)propyl) dimethylsilane	DFSM$_2$	difluoro(3-(2-(2-methoxyethoxy)ethoxy)propyl) methylsilane
LCO	LiCoO$_2$ (lithium cobalt oxide)	LiTFSI	lithium bis (trimethylsulfonyl) imide
LiODFB	lithium oxalydifluoroborate	IL	Ionic liquid
AN1IL-TFSI	IL with allyl group	CEN1IL-TFSI	IL with cynoethyl group
SN1IL	((2-trimethylsililoxyethyl) trimethylammonium bis(trifluoromethanesulf-onyl)imide	LMNO	LiMn$_{1.5}$Ni$_{0.5}$O$_4$
PYR13FSI	N-methyl-N-propyl pyrrolidinium, bis(fluorosulfonyl) imide	PYR13TFSI	N-methyl-N-propyl pyrrolidinium, bis(trifluoromethanesulfonyl) imide
LiFePO$_4$	Lithium iron phosphate	TMP	trimethyl phosphate
NaPF$_6$	sodium hexafluorophosphate	ADN	adiponitrile
LTO	Li$_4$Ti$_5$O$_{12}$	NMC	LiNi$_{1/3}$Co$_{1/3}$Mn$_{1/3}$O$_2$
DMAC	N, N Dimethylacetamide	PFMP	perfluoro-2-methyl-3-pentanone
FS	Fluorocarbon surfactant	F-EPE	1,1,2,2-tetrafluoroethyl-2,2,3,3-tetrafluoropropyl ether
LiBOB	lithium bis(oxalato)borate	GBL	gamma-butyrolactone
FEC	fluoroethylene carbonate	FEMC	3,3,3-fluoroethylmethyl carbonate
HFE	1,1,2,2-tetrafluoroethyl-2′,2′,2′-trifluoroethyl ether	TEP	Triethyl phosphate
DFDEC	di-(2,2,2 trifluoroethyl) carbonate	PC	Propylene carbonate
DMMP	dimethyl methyl-phosphonate	FAP	fluorinated alkyl phosphates
TFEP	tris(2,2,2-trifluoroethyl) phosphate	LiNTF$_2$	lithium bis(trifluoromethane sulphonyl) imide
TMS	tetramethylene sulfone	VC	vinylene carbonate
PEO	poly(ethylene oxide)	PMHS	poly-methyl hydrogen-siloxane
NHC-BF$_3$	1,3-dimethylimidazolidin-2-mm-trifluoroborate	NMC111	LiNi$_{1/3}$Co$_{1/3}$Mn$_{1/3}$O$_2$
NHC-PF$_4$CF$_3$	1,3-dimethylimidazolidin-2-mm-tetrafluorotrifluoromethylphosphate	GPE	gel polymer electrolytes
PVDF	polyvinylidene fluoride	HNT	halloysite nanotube
P(MMA-AN-EA)	poly(methyl methacrylate-acrylonitrile-ethyl acrylate)	EMITFSI	1-ethyl-3-methylimidazolium bis(trifluoromethanesolfonyl) imide
GPE-SN	gel polymer electrolyte containing succinonitrile	GPE-SN-IM	GPE-SN by immersion method
PAN	polyacrylonitrile	PVA	poly(vinyl alcohol)
NIPS	non-solvent induced phase separation	IPN-GPE	Gel polymer electrolyte with interpenetrating polymer network
EMS	ethyl-methyl sulfone	PDLLA	poly(D,L-lactide)
PEGMA	poly(ethylene glycol)methyl ether methacrylate	LAGP	Li$_{1.5}$Al$_{0.5}$Ge$_{1.5}$(PO$_4$)$_3$
ILGPE	ionic liquid gel polymer electrolytes	PEM	polymer electrolyte membrane
PEGBCDMA	polyethylene glycolbiscarbamate dimethacrylate	PAMM	poly(acrylic anhydride-2-methyl-acrylic acid-2-oxirane-ethyl ester-methyl methacrylate)
PCL/SN	Poly(ε-caprolactone)/Succinonitrile	CPE	composite polymer electrolyte
Mg(OH)$_2$	Magnesium hydroxide	PAEKNW	poly (aryl ether ketone) nonwovens
MDPCT	methyl diethyl phospho- noacetate	CETPE	Carbethoxy ethylidene triphenylphosphorane
TFPCT	triethyl2-fluoro-2-phosphonoacetate	Tetra PEG	Tetra-armed poly(ethylene glycol)
NCA	LiNi$_{0.8}$Co$_{0.15}$Al$_{0.05}$O$_2$	PVDF-HFP	polyvinylidenefluoride-hex- afluoropropylene
PBO	poly [benzyl methacrylate-co-oligo(ethylene glycol)ether methacrylate]	c-PPO	cross-linked poly[dimethyl-p- vinyl benzyl phosphonate-co-oligo (ethylene glycol) meth acrylate] co-polymer (c-PPO)
DTYP	diethyl(thiophen-2-ylmethyl) phosphonate	PFN	Fluorinated phosphazene derivative, ethoxy-(pentafluoro)-cyclotriphosphazene
PFPN	ethoxy(pentafluoro) cyclotriphosphazene	EEEP	Poly[bis-(ethoxyethoxyethoxy)phosphazene]

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