# Utilizing membrane technologies in advancing the recycling of spent lithium-ion batteries using green electrochemical method – A review

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**Abstract.** The demand for lithium (Li) resources is soaring due to the widespread production (or consumption) of electronic products such as mobile and electronic devices, (or laptops, tablets, and home-appliances goods in accommodating current global lifestyle). The other lithium-ion batteries (LiBs) applications include electric vehicles, solar panels, wind turbines, and electric toys. The increasing demand for Li, while driving the economic progress of the industry, is putting a strain on the resource reserves. Therefore, the production industry is promptly searching for an efficient spent LiBs recycling process to counterbalance the highly sought-after element. Current Li recycling systems, in which extraction and recovery are typically accomplished by hydrometallurgical processes, have a significant impact on the environment, are energy-intensive, and necessitate vast operational capacities. Recently,  electrochemical methods are seen as

sustainable and green approaches to Li production. The use of membrane materials for Li recovery together with electrochemical processes provides a means to reduce energy consumption and scale up the spent LiBs recycling. In this overview, Li recovery technologies through pyrometallurgy, hydrometallurgy and green electrochemical extraction are explored along with their benefits and drawbacks. Recent advances in membrane materials selection that lead to significant improvement in Li production are also discussed.

**Introduction to Lithium-ion Battery Recycling**
Lithium-ion batteries (LiBs), as the most significant candidate for energy storage devices, have quickly occupied the global electrical consumer market due to their relatively high energy density, advanced operating voltage, and lack of a memory effect [1]. Statistics by GlobeNewswire reported that the global market for LiBs is forecasted to increase from USD 41.1 billion in 2021 to USD 116.1 billion by 2030, where the compound annual growth rate (CAGR) is projected to grow by 12.3% from 2021 to 2030. Factors such as declining LiBs prices and the high demand for batteries from the automotive industry are expected to drive the battery market during the forecast period. The increasing need for a 'battery material' supply has led to the continuous search for Li global reserves. Hence, it is expected that the amount of Li in the world is shrinking rapidly and will not be able to meet future demand [2]. Therefore, there is a need to develop or improve the current Li production and recovery system to ensure supply chain resiliency.

There are three (3) main methods that have been developed to recover Li from waste LiBs. They are electrochemical extraction, pyrometallurgy, and hydrometallurgy [3]. Fig. 1 shows the comparison of the three Li recycling methods based on energy efficiency, chemical cost, recycling efficiency, simplicity, environmental friendliness, and scale-up state. As shown in the figure, by and large, the electrochemical extraction method is a more environmentally benign option than pyrometallurgy and hydrothermal processes [3–6]. The electrochemical extraction can be operated at room temperature, produce less chemical waste, and is more energy efficient. Furthermore, it can yield Li with >99% purity. Meanwhile, the other two methods need to be run at the expense of a higher temperature setup and have the potential to cause acid, volatile compounds, and organic waste pollution. Nonetheless, the recycling efficiency of Li for the electrochemical extraction method is only 75% to 98% when the conductive ceramic solid electrolyte is utilized as the membrane separator. In addition, the electrochemical extraction process is currently at the proof-of-concept stage and is yet to be commercialized despite its advantages. Complex electrochemical setup and the fabrication of Li separators remain a major concern. Such limitations bring more value to Li mining, underscoring the significance of Li recycling.

The purpose of this paper is to serve as a reference for researchers, scientists, engineers, and technologists working to improve spent LiB recycling and recovery processes in the future. The review also details the processes, benefits, and downsides of the membrane materials for Li recycling that are already in use. The direction for future Li recycling technology research and development is suggested.
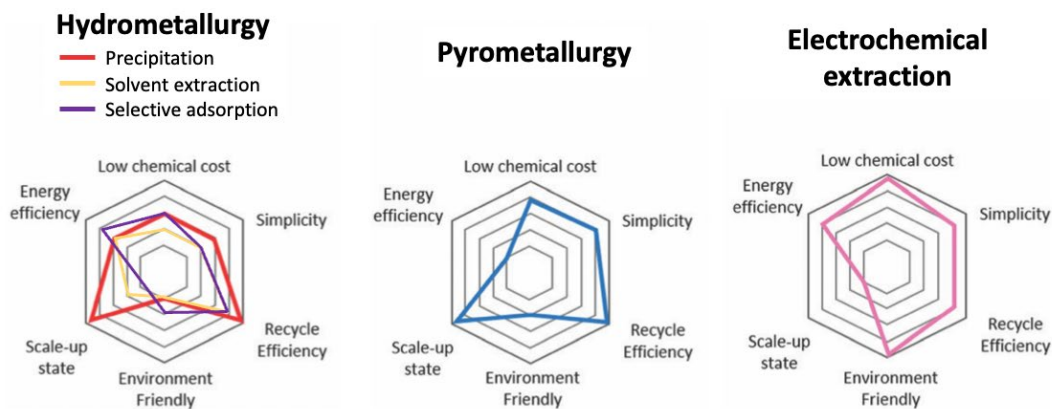
*Fig. 1. Comparison of lithium recycling methods. Adapted from Bae et al. [3].*

**Lithium Recovery Technologies**

The efficiency of Li extraction is a major consideration in selecting recovery technologies. Technically, the metal-extraction process is employed to transform the solid metals found in used LiBs into their alloy form or solution state. Techniques like pyrometallurgy, hydrometallurgy, and electrochemical are commonly used during the extraction of Li in active materials (see Fig. 2). However, towards the sustainable environment and life, the most preferred Li ion recycling method globally is the method which could support and stand for the green technology concept.

The 'green technology' concept which is emphasized for LiBs recycling methods in this article can consist of several temperaments. The most important ones are the characteristics which are included within recycling process such as waste prevention especially into the earth, energy usage reduction which can be translated into application of ambient temperature and pressure, inclusion of synthetic methods or chemical products which are designed to significantly reducing toxicity, inclusion of synthetic methods that able to ensure all materials incorporated within the process to be fully utilized and the minimization of auxiliary substances. More on that, the recycling process should prevent the application of stoichiometric reagents and replace it with catalytic reagents after careful consideration. All substances included in the recycling process should be able to minimize the chemical or physical accidents. Besides, the characteristics which should be included in the raw materials selections is that the materials should exist in renewable form [6]. The following sections discuss each extraction method and its efficiency as reported in the literature.
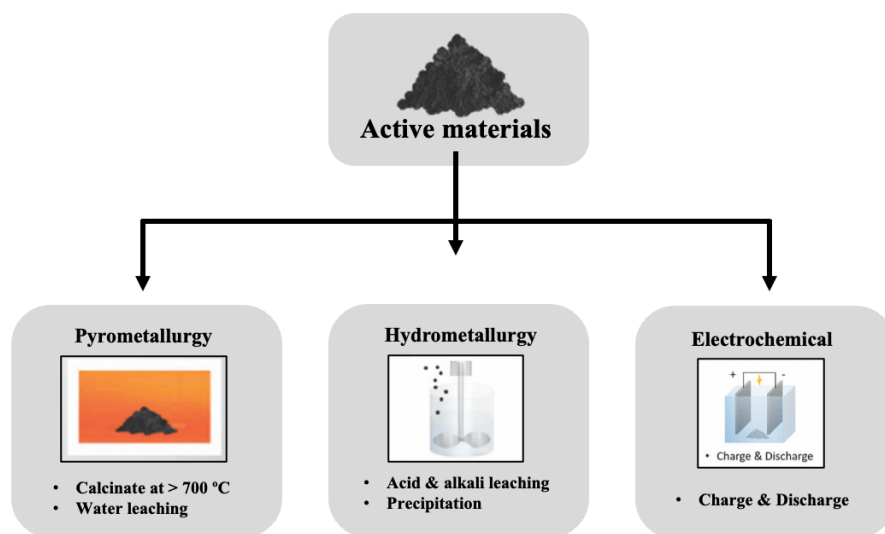
*Fig. 2. Schematic diagram of three (3) main recycling stages and methods from active materials.*

*Pyrometallurgy*

Much research highlights that pyrometallurgical is a prevalent and practical method for metal extraction, including for LiBs recycling. To recycle old batteries, for instance, a Umicore process could be used [8]. During the process, the active materials are melted down in a furnace in a smelting process. Compounds like polymers, organic solvents, and graphite in the batteries are decomposed while metal components are reduced and converted to alloys. The resulting alloys are further treated using sulfuric acid ($H_2SO_4$) as the leaching agent and solvent extraction to obtain cobalt oxides (CoO) and nickel hydroxide ($Ni(OH)_2$) [9]. Although this process does not require pre-treatment, it could significantly impact the recovery of Li compositions [10]. Due to this issue, researchers have proposed a unique approach that combines pyrometallurgy and hydrometallurgy to recycle valuable metals from wasted LiBs, intending to minimize Li loss during recovery.

In a study, Georgi et al. [11] were able to salvage precious metals from used LiBs by employing a reduction smelting technique. Important metals, including Fe, Co, Ni, and Mn, were transformed into alloys. However, Li went into slag or dust throughout the process. $H_2SO_4$ leaching was used to further isolate Li. Further, Werner et al. [12] proposed a method to evaporate Li from used LiBs using a combination of vacuum evaporation and selective carrier gas evaporation at high temperatures. But, the process temperature was higher than 1400 °C, which requires substantial energy usage. Another study by Peng et al. [13] reported a novel approach to recycling wasted LiBs, roasting them at a low temperature (25 °C to 70 °C) in an argon environment (see mechanism illustrated in Fig. 3), then extracting the $Li_2CO_3$ using water leaching. After the crystallization and leaching processes, the Li concentration in the leaching solution reached 4.36 g/L. Generally, the pyrometallurgy process to recover important metals from spent LiBs is straightforward, but it is not eco-friendly because of the high energy consumption and the potential secondary pollution [14,15] which disobeyed the green technology concept that should aimed to reduce energy consumption and prevent substance or products wastes. In addition, Li loss during the recycling process is also significant and needs to be addressed in future studies. Table 1 summarised the past reviews on Li extraction using pyrometallurgy.
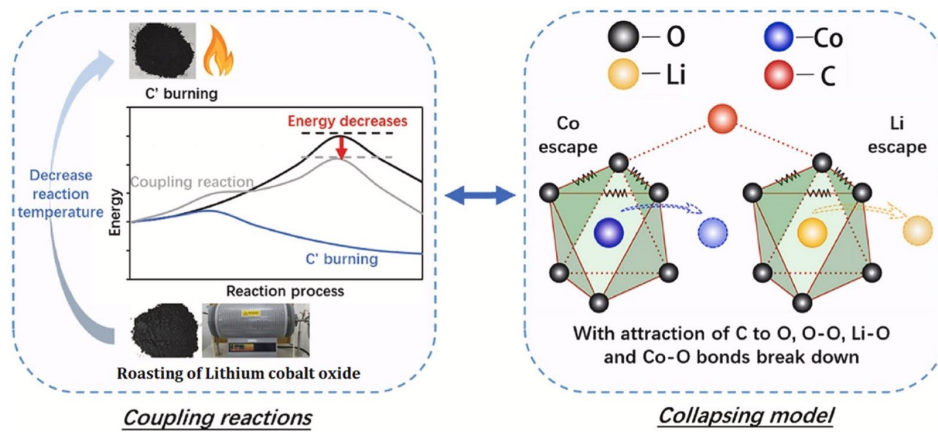
*Fig. 3. Illustration mechanism of coupling reaction and collapsing model in carbothermic reduction roasting. During this process, carbothermic reaction facilitates the reduction by destroying oxygen octahedrons (O) in the metal oxide crystal formations. The collapse of the O octahedrons allows Li and Co to escape at ease, driving the reaction forward. Adapted from Peng et al. [13].*

*Table 1. List of past reviews and their research summary on Li extraction using pyrometallurgy.*

| Method | Steps | Summary | Ref. |
|---|---|---|---|
| Pyrometallurgy | Umicore process | The active materials were melted down in a furnace and converted into alloys. Further recovery of Li was achieved using $H_2SO_4$ as the leaching agent | [8] |
| | Reduction smelting | Li went into slag or dust throughout the process and the use of $H_2SO_4$ as the leaching agent promotes Li's isolation | [11] |
| | Vacuum evaporation and selective gas carrier | Li extraction was recovered through evaporation process from the used LiBs at high temperatures up to 1400 °C | [12] |
| | Roasting and water leaching | Roasting the used LiBs at low temperatures of 25 °C to 70 °C in an argon environment, thus extracting $Li_2CO_3$ using water leaching | [13] |

*Hydrometallurgy*

Hydrometallurgy is the most widely used laboratory metal recovery technique and it is on the brink of commercialization. The method secures high recovery rates with few supplementary equipment [16,17]. First, acids and bases are used to ionize Li in pre-treated active materials. Following that, Li is recovered from the resulting $Li^+$ solutions by leaching of acids. Sulfuric acid, hydrochloric acid, citric acid, and nitric acid are examples of acids that were employed [18,19]. The leaching efficiency was improved by applying redox reactions with hydrogen peroxide ($H_2O_2$). Due to its high reducibility and low toxicity, the redox agent could speed up the leaching procedure. Nevertheless, a low-pH acid has the potential to release $Cl_2$ and $NO_2$, which have negative impacts on the environment (soil and air pollution) [20]. Therefore, it should be avoided if possible. In a past study, Zhang et al. [21] utilized biodegradable tritrifluoroacetic acid (TFA) and trichloroacetic

acid (TCA) as the leaching agent, while $H_2O_2$ as the reducing agent (see Fig. 4 (a)). As a result, the Li leaching efficiencies reached 99.7%, under optimal conditions.

Another study by Chen et al. [22] found that a 96% leaching rate of Li extraction could be achieved by employing multiple processes including acid leaching, alkali leaching, solvent extraction, and chemical precipitation. An $H_2SO_4$ was used as a leaching agent and $H_2O_2$ as a reducing agent. Li et al. [23] devised a three-step procedure that includes ultrasonic cleaning, roasting, and organic acid leaching. A 1.25 mol/L of ascorbic acid was used as both the leaching agent and the reducing agent (25 g/L ratios), resulting in 98.5% of Li leaching rates. Chen et al. [24] invented a cost-effective method for recycling precious metals from used LiBs (see Fig. 4 (b)). In the extraction process, citric acid was selected as the leaching agent, and D-glucose was employed as the reducing agent to dissolve the spent cathode material. The dissolution reaction resulted in approximately 99% leaching efficiency of the Li. Furthermore, Chen et al. [25] reported that $LiCoO_2$ could be extracted into a precipitate and a Li solution using tartaric acid, which demonstrated a 97% recovery performance of Li. Overall, the recycling of Li via hydrometallurgy had an utmost efficiency of up to ~98%. However, this process necessitates a huge amount of acids and bases as leaching agents which does not come cheap. This factor alone already diverted from the green technology concept which should reduce toxicity and substances should have less risk towards safety and health. Moreover, there may be substantial secondary costs to dispose the used solutions and disobey the green technology concept which should prevent waste substances or products. Not to forget that environmental pollution still emerged as an issue in hydrometallurgy. Table 2 summarised the past reviews on Li extraction using hydrometallurgy.
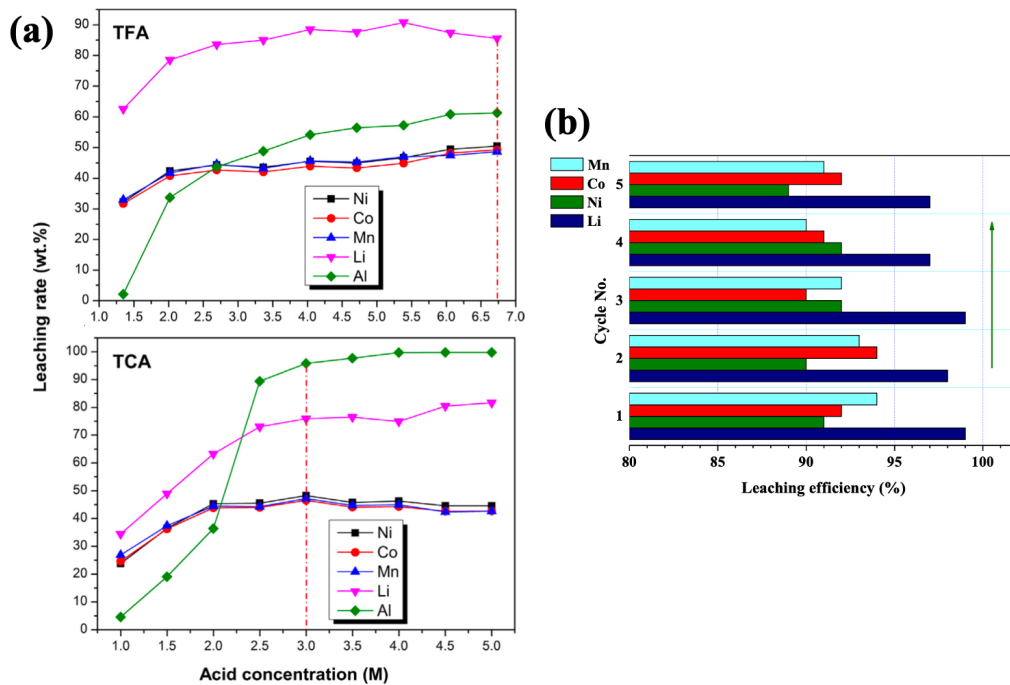


Fig. 4. The effects of biodegradable acids (TFA and TCA) on their leaching rates of metals under optimal conditions (4 vol% $H_2O_2$, 60 °C temperature in 30 min) [19]. (b) Leaching efficiency of citric acid to precious metals; Mn, Co, Ni, and Li under conditions of 1.5 mol/L citric acid concentration, 80 °C temperature, and 120 min reaction time [22].

*Table 2. List of past reviews and their research summary on Li extraction using hydrometallurgy.*

| Method | Summary | Ref. |
|---|---|---|
| Hydrometallurgy | The uses of acid and bases *e.g.* HCl as leaching agents could be used to ionize $Li^+$. Moreover, the additional redox reaction with $H_2O_2$ could significantly improve the leaching efficiency. However, low pH acid could impact the release of $Cl_2$ and $NO_2$ which promotes soil and water pollution. | [16,17] |
| | 95% leaching rate of Li extraction could be achieved by employing multi-steps processes including acid and alkali leaching, solvent extraction and chemical precipitation. | [22] |
| | 98.5% leaching rate of Li extraction was achieved in three steps procedure including ultrasonic cleaning, roasting and organic acid leaching. | [23] |
| | A cost-effective utilising citric acid and D-glucose as the leaching and reducing agent resulted in approximately 99% leaching efficiency of Li. | [24] |
| | $LiCoO_2$ could be extracted into precipitates using tartaric acid, which demonstrated 97% leaching efficiency. | [25] |

*Electrochemical extraction*

Mainly, based on multiple dimensions, the electrochemical extraction method is considered more environmentally friendly than the pyrometallurgy and hydrothermal processes [26, 27]. It can be run at room temperature, does not produce a lot of chemical waste, and does not require much energy to operate. These characteristics made this electrochemical extraction method into the green technology list and tick all the necessary green characteristics. Furthermore, it can yield Li up to 99% purity [6]. The schematic diagram of the electrochemical setup is illustrated in Fig. 5 (a). However, the Li recycling effectiveness for the electrochemical extraction approach using an LTAP ceramic solid electrolyte as the membrane to recover Li from pre-treated active materials is only 75%. Also, despite its benefits, the electrochemical extraction study has only been conducted on a lab scale and has not yet been commercialized.

The electrochemical Li extraction system is capable of extracting Li via the charging and discharging mechanism [28]. Kanoh et al. [29] in their early study performed the first electrochemical capture of Li cations from a solution in battery material. Li intercalation, the most common way for Li-ion batteries to recharge, formed the basis of the capture process. They generated oxygen or hydrogen at a manganese oxide ($MnO_2$) working electrode and a platinum (Pt) wire counter electrode. However, this technique has some drawbacks due to the expensive process of water splitting reaction undergoing simultaneously with the Li recovery [28]. Following this, Pasta et al. [30] proposed an alternative method to capture and release Li ions and anions. The phrase "*electrochemical ion pumping*" was recently used to describe this innovation [30–32]. Since electrochemical ion pumping only involves breaking and production of chemical bonds, it requires (OR reasonably/rationally consume) less energy.

Electrodialysis is another method used in electrochemistry to extract Li from various materials (brines and seawater) [2,32]. This method relies on a selective membrane that allows only Li$^+$ ions to pass through. The primary benefit this (OR of the) technique is the voltage which provides the driving force for capturing Li. Therefore, no other chemical species is required to replenish the active material. To date, approximately 95% to 97% purity extraction was reported by using this method. Other works of literature reported that this method requires less time consumption [34, 35]. The increased capture rate of Li is another direct result of applying current to the electrode in electrodialysis [36]. The charging and discharging process (see Fig. 5 (b)) are the basis needed for the electrochemical method with no further chemicals or heat involved. It is worth noticing that this method is orchestrated as a green Li recycling.

Moreover, Li purity can be extracted through a designed solid electrolyte that enables only Li to flow through. However, this technique has only been validated on a lab scale, whilst the properties of the membrane extractor are important to facilitate the movement of Li-ions and recover Li with high purity. Several studies have been conducted to upgrade the properties of the membrane. Membrane extractors such as solid polymer electrolytes (SPE), gel polymer electrolytes (GPE), and composite polymer electrolytes (CPE) were designed to increase the efficiency of recycling Li purity [37–39]. These membrane strategies are explained in the following sections which include their functionalities, and an ongoing process in this area. Although this method could exhibit the highest efficiency (97.20% to 99.01%) of Li recovery compared to hydrometallurgy and pyrometallurgy processes, the drawbacks such as complex electrochemical setup and the selection of the membrane separator are crucial. Table 3 summarised the past reviews on Li extraction using electrochemical extraction.
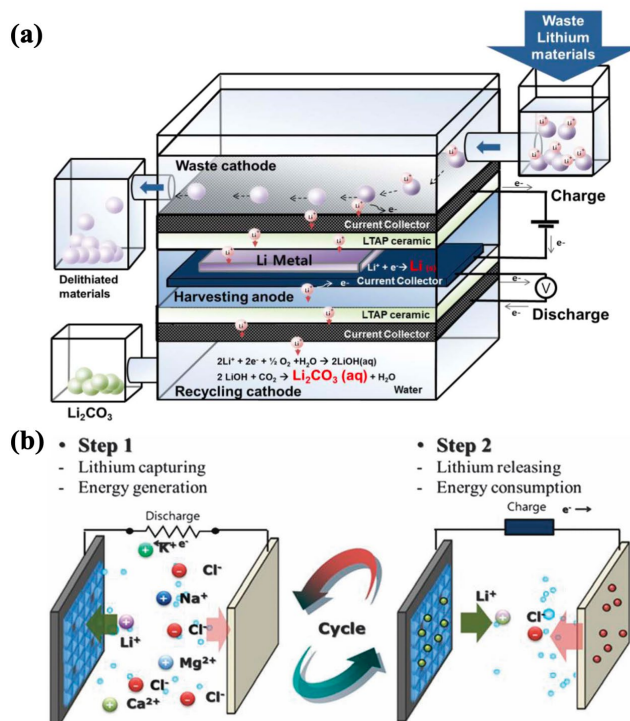


*Fig. 5. (a) A schematic diagram of electrochemical setup in recycling Li. During charging, Li materials in the waste cathode compartment migrate to the harvesting anode compartment, where they combine to generate Li metal. Following this, by reacting with $H_2O$, $O_2$, and $CO_2$, the recycling cathode compartment generates $Li_2CO_3$ or LiOH during system discharge [5]. (b) In step 1, the discharging process involves capturing Li ions in the source water, and in step 2, the releasing Li process occurs in the reservoir solution [36].*

*Table 3. List of past reviews and their research summary on Li extraction using the electrochemical method.*

| Method | Summary | Ref. |
|---|---|---|
| Electrochemical extraction | A 75% Li extraction could be achieved via LTAP ceramic solid electrolyte as the membrane separator | [6] |
| | Li extraction through the charging and discharging mechanism in electrochemical setup. The separator was used to capture Li, while MnO2 and Pt as the working electrode and counter electrode, respectively | [28] |
| | Electrochemical ion pumping was discovered to capture and release Li ions. This method requires less energy consumption | [30] |
| | A 95% to 97% of Li extraction was considered in the electrodialysis step utilising brines and seawater separators | [2,32] |
| | A 97.20% to 99.01% of Li purity could be extracted through a designed SPE, GPE, and CPE as membrane separator | [37–40] |

**Membrane Polymeric Materials for Lithium-ion Recycling in Green Electrochemical Method**

In the electrochemical method, the ionic conductivity of membrane materials is the prime factor in recycling Li as it will ensure an efficient migration and high purity of Li ions to pass through the membrane. Previously, high Li ion conductivity was provided by the traditional liquid electrolytes, but safety concerns are raised due to the flammability of organic solvents, like ethylene carbonate and dimethyl carbonate in the electrochemical medium [41,42]. As a result, the development of polymer-based, solid polymer electrolytes (SPEs) has been the focus of study for quite some time. The rate of electrode/electrolyte interface deformation is reduced, leading to improved thermal, mechanical, and electrochemical stability [43,44]. While these polymer electrolytes meet all of the necessary safety standards, their ionic conductivity is typically lower than that of liquid electrolytes. In a past study, Johari et al. [45] reported polyethylene oxide (PEO) based Li/SPE membrane possessed low ionic conductivity, $\sigma$, at approximately $\sim 10^{-7}$ S/cm at room temperature. This phenomenon could be explained mainly due to the crystalline phase of polymers, which restricted their wide utilization [46,47]. Therefore, a lot of materials and methods were explored to reduce the glass transition temperature ($T_g$) and improve the conductive network of SPEs.

SPEs consist of a primarily polymeric matrix with one or more fillers, which may be ceramic, Li salts, or ionic liquids. Generally, the ionic conductivity of an SPE is a key factor, therefore fillers are often added to the material to enhance its conductivity. The introduction of those fillers could decrease the polymer's crystallinity, which significantly improves ion migration and ionic conductivity. Poly(ethylene oxide) (PEO), poly(vinylidene fluoride) (PVDF) and its copolymers, poly(ethylene glycol) (PEG), poly(acrylonitrile) (PAN), and poly(ethylene carbonate) (PECO$_3$) are the most used (OR utilized/applied) polymer matrices for SPEs [48–50]. Li salts, ionic liquids, composite polymers, and interpenetrating network polymers are all examples of modified membranes. The following section discussed the current findings on the abovementioned membrane materials, their potentials, and their drawbacks in recycling Li in the green electrochemical method.

*SPEs-based Li salts and ionic liquid*

In recent studies, Li salts such as lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) and lithium perchlorate (LiClO$_4$) have been shown to be the most effective active fillers for SPEs' membrane [51,52]. The ionic conductivity and the number of charge carriers both rise (OR grow/increase/raise) directly (OR instantly/promptly) as a result of (OR resulted from or due to) the presence of these salts. Moreover, ionic liquids like 1-ethyl-3- methylimidazolium bis(trifluoromethylsulfonyl)imide (EMIM)(TFSI), 1-butyl-3-methylimidazolium chloride (BMIM)(Cl) and lithium tetrafluoroborate (LiBF$_4$), lithium hexafluorophosphate (LiPF$_6$) could also improve the ionic conductivity of SPEs [53,55]. According to Yoon et al. [53], the ionic liquid could influence the ion-dipole interactions between ions and the polymer matrix. Therefore, these interactions promote Li salt dissociation and boost the number and mobility of Li-ion transfers. In the end, it could produce a high yield of Li recovery.

Mendes et al. [56] designed polyurethane acrylate (PUA) and bis(trifluoromethanesulfonyl) imide (LiTFSI) salts as SPE membranes. The introduction of LiTFSI significantly modified the surface of SPEs. The ionic conductivity of UV photocurable in 30 wt% LiTFSI content reached a maximum of 0.0032 mS/cm at ambient temperature. In addition, the limited oxidative stability and poor interfacial charge transport of existing SPEs can be ameliorated by employing ether-based electrolytes with the presence of aluminum fluoride (AlF$_3$) [57]. Incorporating PEO into a matrix containing lithium trifluoromethyl silane (LiTFSI) resulted in the creation of an SPE with impressive flame retardant capacity, enhanced mechanical stability, and a wide electrochemical stability window of up to 4.6 V. In addition, their study also embarks on the addition of sodium alginate (SA) to improve the mechanical properties of SPE and the safety of the membrane through the nanofiber structure [58]. Figs. 6 (a and b) illustrate the physical appearance and the morphological images of the membrane.

In a previous study by Kale et al. [59], a more complicated composite of poly(ethylene oxide) monomethacrylate, PEGMA, and cellulose filled with LiTFSI and 1-butyl-1-methylpyrrolidinium bis(trifluoromethanesulfonyl)imide (Pyr14TFSI) (see schematic diagram in Fig. 6 (c)) also (OR have) achieved a good cycling stability at room temperature with an average transference number of 0.43 and electrochemical stability window up to 5 V. Further, the membrane does offer/present/provide flexible attribute/characteristic as it does not succumb to bending and even twisting (Figs. 6 (d and e)). The crystallinity of the SPE can be lowered by including several Li salts into a PMMA matrix grafted with natural rubber, which in turn increases the Li transference number and, by extension, the ionic conductivity of the material. However, the use of this type of ionic liquid and Li salts-based SPEs has a drawback, which is the toxicity impact due to the possibility of release into the soil or water courses, where they could become persistent pollutants and pose environmental risks. Table 4 depicts the natural polymers and their fillers that have been the basis for recent developments in SPEs membranes.
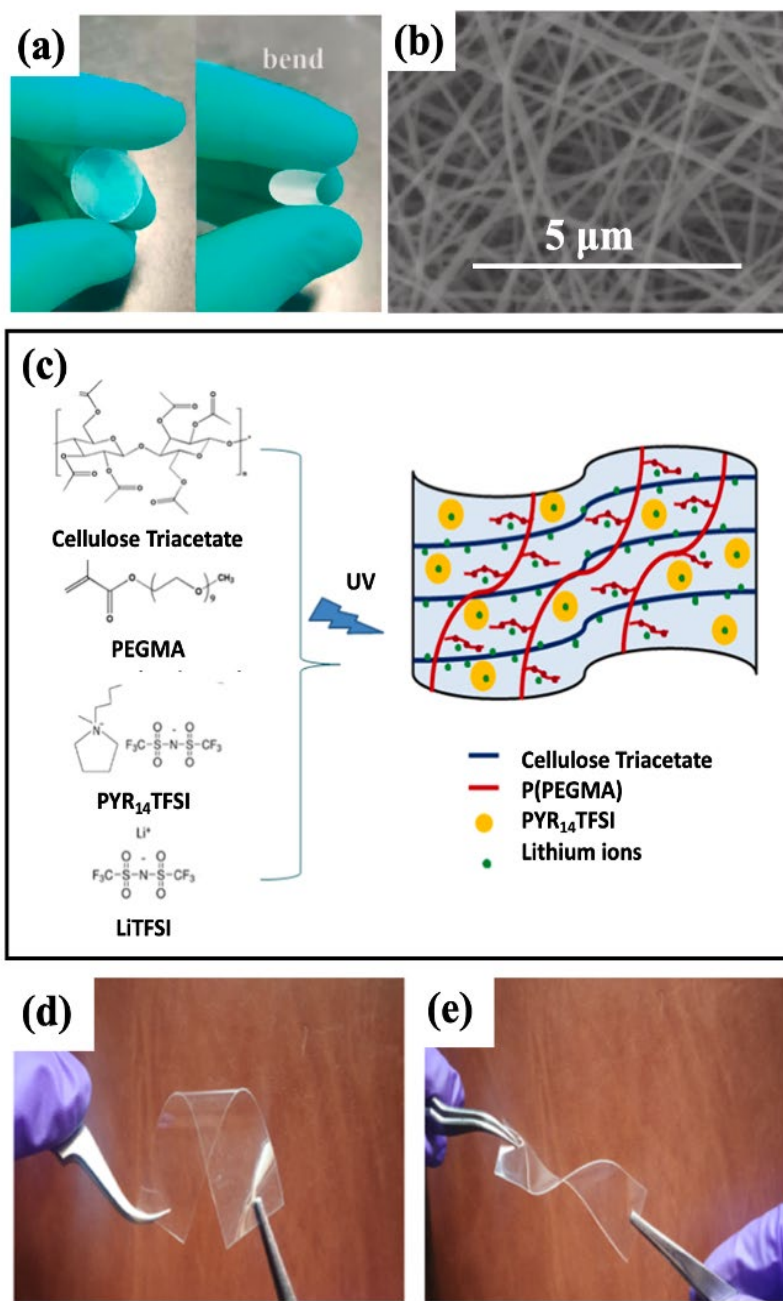
*Fig. 6. (a) A free-standing film of PEO/SA-SPE membrane, (b) which is randomly pilled at ~170 nm observed by SEM images [56]. This is believed as an ideal 3D framework for structural support of membranes. (c) Li coordination site accessibility and polymer synthesis reaction scheme. (d and e) SPE membranes that can stand on their own, are transparent, and flexible in bending and twisting [59].*

*Table 4. Natural polymers and their fillers have been the basis for recent developments in SPEs membranes.*

| Polymer Matrix | Fillers | Ionic conductivity (S/cm) | Reference |
|---|---|---|---|
| PMMA/natural rubber | LiBF | $1.89 \times 10^{-6}$ | [60] |
| Cellulose | $LiPF_6$ | $2.71 \times 10^{-2}$ | [61] |
| Triacetate/PEGMA | LiTFSI | $5.24 \times 10^{-3}$ | [59] |
| PEO/SA | LiTFSI | $\sim10^{-4}$ | [58] |

*Composite polymer electrolytes (CPEs) membrane*

In a recent study, Kim et al. [62] utilized the UV radiation approach to increase the mechanical strength of the PEO membrane. It is clear that the polymer can be solidified following UV radiation in corresponds with trimethylolpropane ethoxylate triacrylate (ETPTA) ceramic filler included in the precursor solution. This resulted in the production of a freestanding membrane. At room temperature, the ionic conductivity can reach as high as $3.3 \times 10^3$ S/cm with a 0.76 $Li^+$ transference number. Din et al. [63] created the CPEs with varied structures, such as mixes of PEO with ceramic particles, ceramic networks, and ceramic layers. The 50 to 70 μm thick membrane was created when the nanowire of $Li_7La_3Zr_2O_{12}$ (LLZO) was combined with PEO containing $LiClO_4$ salt. The maximum ionic conductivity of $4.42 \times 10^4$ S/cm was achieved with the addition of 52.5% weight of LLZO. Fig. 7 (a) shows the filler particles of LLZO are packed densely throughout the polymer matrix. By introducing the filler, the PEO undergoes a change in crystallinity that is conducive to high chain mobility, and, by extension, high Li-ion conductivities were produced inside the polymer matrix.

High ionic conductivities could be observed in CPEs including well-aligned conducting nanowires by varying their orientations [64]. Figs. 7 (b-e) illustrates a comparison of the Li conductive pathway in a different alignment. Well-aligned nanowires exhibit $6.05 \times 10^5$ S/cm at 30 ºC conductivity which is 10 times higher than that of their randomly-dispersed counterparts. The calculated surface conductivity of the nanowires at 30 ºC is $1.26 \times 10^2$ S/cm, which is quite close to the ion conduction in liquid electrolytes. A rapid ion conduction channel without surface-to-surface connections is responsible for the notable conductivity enhancement observed in nanowires. The addition of the nanowires also enhances the polymer electrolytes' long-term conductivity stability. Solid electrolytes with in-plane orientation could also pave the way for more efficient Li migration in the membrane. Li et al. [64] developed free-standing nanofibers based on a 3D $Li_{0.35}La_{0.55}TiO_3$ (LLTO) framework. In order to prevent the Li-ion route from being blocked by clumps of ceramic nanoparticles, a 3D framework may be necessary. Their study concluded that the LLTO 3D framework is superior to one made with LLTO nanoparticles. At room temperature, the conductivity reaches its maximum of $8.8 \times 10^5$ S/cm. According to Feng et al. [65], issues like brittleness (poor mechanical properties) and less compatibility with ceramic-polymer adhesion could diminish the Li recovery process. More previous studies following the types of CPEs and their respective conductivity are summarised in Table 5.
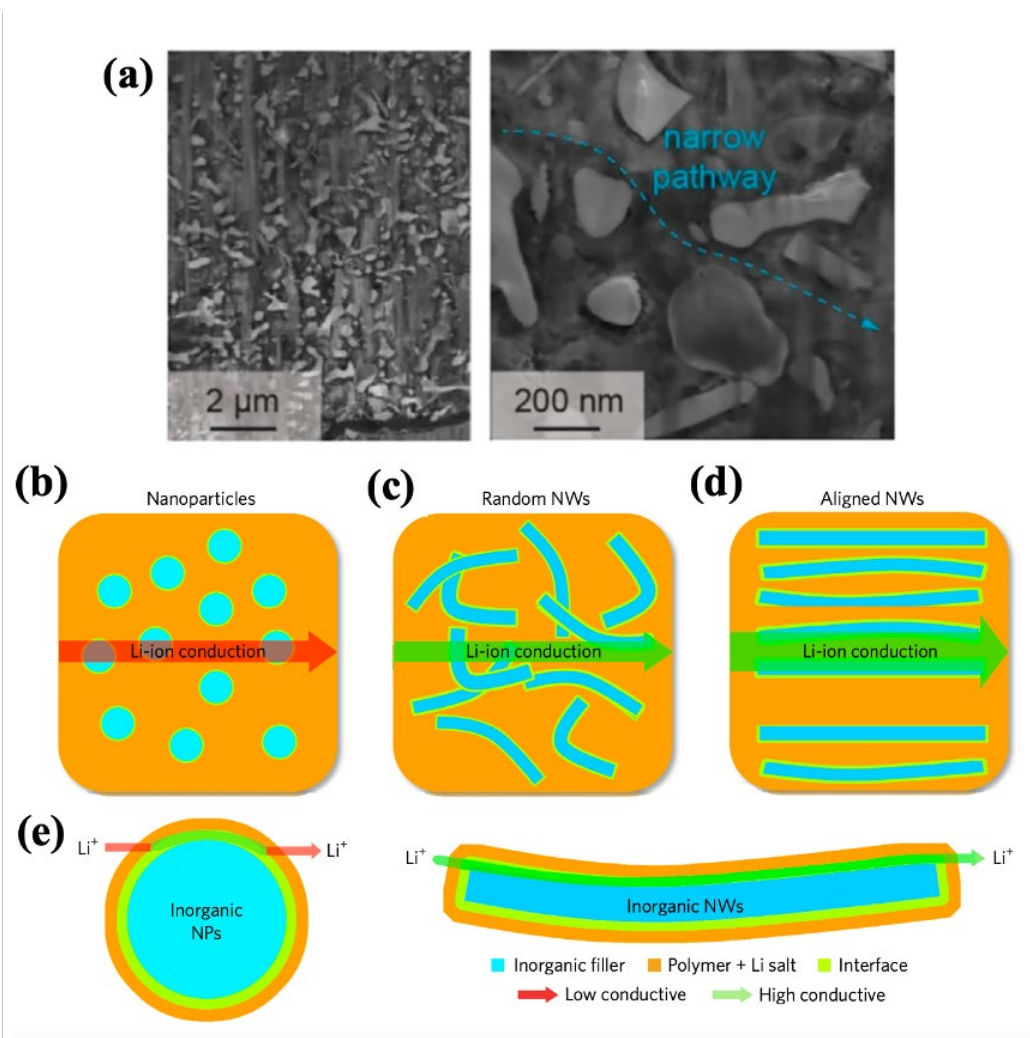
*Fig. 7. (a) SEM images of the cross-sectional area of PEO consisting of 20 wt% nanowires LLZO [61]. The Li-ion conduction routes in CPEs contain (b) nanoparticles, (c) random nanowires, and (d) aligned nanowires. Unpredictable nanowires may provide a more consistent rapid conduction pathway for Li ions than isolated nanoparticles. The crossing junctions do not exist in aligned nanowires, which is a major advantage over random nanowires. (e) Li-ion conduction occurs along the surface of inorganic nanoparticles and nanowires [64].*

*Table 5. Types of ceramic fillers in their electrochemical performances in CPEs.*

| Structural type | Ceramic filler | Polymer host | Ionic conductivity (S/cm) | Reference |
|---|---|---|---|---|
| Nanoparticle | $Li_{6.75}La_3Zr_{1.75}Nb_{0.25}O_{12}$ | PVDF/LiClO$_4$ | $9.20 \times 10^{-5}$ | [67] |
| | LAGP | PET/LiTFSI | $7.78 \times 10^{-5}$ | [68] |
| | LATP | PEG/PEO/LiClO$_4$ | $5.20 \times 10^{-5}$ | [69] |
| Nanowire | $Li_{6.4}La_3Zr_2Al_{0.2}O_{12}$ | PEO/LiTFSI | $2.50 \times 10^{-4}$ | [70] |
| | $Li_{6.75}La_3Zr_{1.75}Nb_{0.25}O_{12}$ | LiClO$_4$ | $2.20 \times 10^{-5}$ | [71] |
| | LLZO | PVDF/LiTFSI | $9.50 \times 10^{-4}$ | [72] |
| 3D framework | $Li_{6.28}La_3Zr_2Al_{0.24}O_{12}$ | LiTFSI | $8.50 \times 10^{-5}$ | [73] |
| | $Li_{6.4}La_3Zr_2Al_{0.2}O_{12}$ | LiTFSI | $1.80 \times 10^{-5}$ | [74] |
| | LLZO | PEO/LiTFSI | $1.14 \times 10^{-4}$ | [75] |

*Interpenetrating network polymer (IPNs) membrane*

The combination of CPEs/SPEs with IPNs is an innovative way to improve the efficiency of Li's recovery performance. Generally, IPNs are made up of two or more cross-linked polymers that are not bonded to one another [76]. Reviews have found that IPNs can improve ion conductivities and mechanical properties by reducing their crystallinity [77]. A past study by Zeng et al. [78] developed the IPN-based poly(ether-acrylate) that possessed $2.2 \times 10^4$ S/cm at room temperature with a low glass transition temperature at 64.2 ºC. The newly ventured electronically conductive polymers (CPs) namely poly(3,4-ethylene dioxythiophene) (PEDOT) have taken place in designing the basic electrode of supercapacitors [79]. In this semi-IPN framework, it is possible to create new interconnecting materials or membrane separators that are based on CPs [80].

This strategy was designed to replace the older method, which uses organic liquid electrolytes. These CPs are chosen due to their unique IPNs that are capable of decreasing the crystallinity and therefore enhancing ionic conductivities and mechanical properties. IPN structure in SPE-CPs could be compromised into multiple networks which are at least partially interlaced on a polymer scale but not covalently bonded to each other. Other characteristics such as (1) good thermal stability, (2) high electrolyte wettability, (3) redox-active separator, (4) electron-rich groups, (5) fibrous structure, and (6) capacity-enhancing could drive the ion exchange through the SPE-CP membrane. When applied as the membrane separator for Li extraction, the electronic nature of CPs should be eliminated to avoid the risk of a short circuit between positive and negative electrodes. To solve the issue, Wang et al. [81] embarked on the electrochemical overoxidation of CPs, prior to their incorporation into the SPE membrane. It offers a loss of electronic conductivity, the formation of carbonyl groups that can attract cations, and hinders the diffusion of anions through the membrane. Based on this, the electron-rich groups are introduced into the conjugated CP's structure and converted into a rapid ion-exchange membrane. This newly emerged technique aims to enhance and broaden the useful features of original SPE compounds.

The CP-based SPEs could offer (OR provide) ionic conductivity that exceeds $10^{-5}$ S/cm, facilitating the migration of Li ions through the membrane. Besides, $Li^+$ is typically less dynamic than its anionic counterpart since its motion is coupled with the lewis basic sites of the polymer matrix. The ratio of migrating $Li^+$ to all migrating ions, including anions in the electrolyte is defined as the Lithium Ion Transference Number (LITN) [82]. During charge/discharge cycling, only migrating $Li^+$ contributes to the performance of the battery. Overall, the ideal LiTN is equal to 1, which is achievable by CP. In terms of physical properties, CPs can offer great fiber orientation, as their porosity is tuneable and can be designed accordingly. This allows the acceleration mechanism of $Li^+$ during the extraction process. Fong et al. [79] developed the semi-IPNs-based PEDOT. The material's shape enables these enhancements in specific capacitance, as depicted schematically in Fig. 8 (a). The ionically PEDOT in post-overoxidation provides an ion reservoir throughout the electrode, making pseudocapacitive polymer available for charge storage processes even inside the bulk of the material. This technology improves the material use efficiency of an electrically CP without requiring sophisticated production procedures or compromising mechanical stability. In addition, the flexible framework structure characteristic of the ionically conductive matrix offer multiple advantages including; (1) able to accommodate the volumetric variations associated with ion intercalation/deintercalation in the pseudocapacitive polymer, (2) decrease mechanical stress in the electrode and (3) produce high cycling stability.
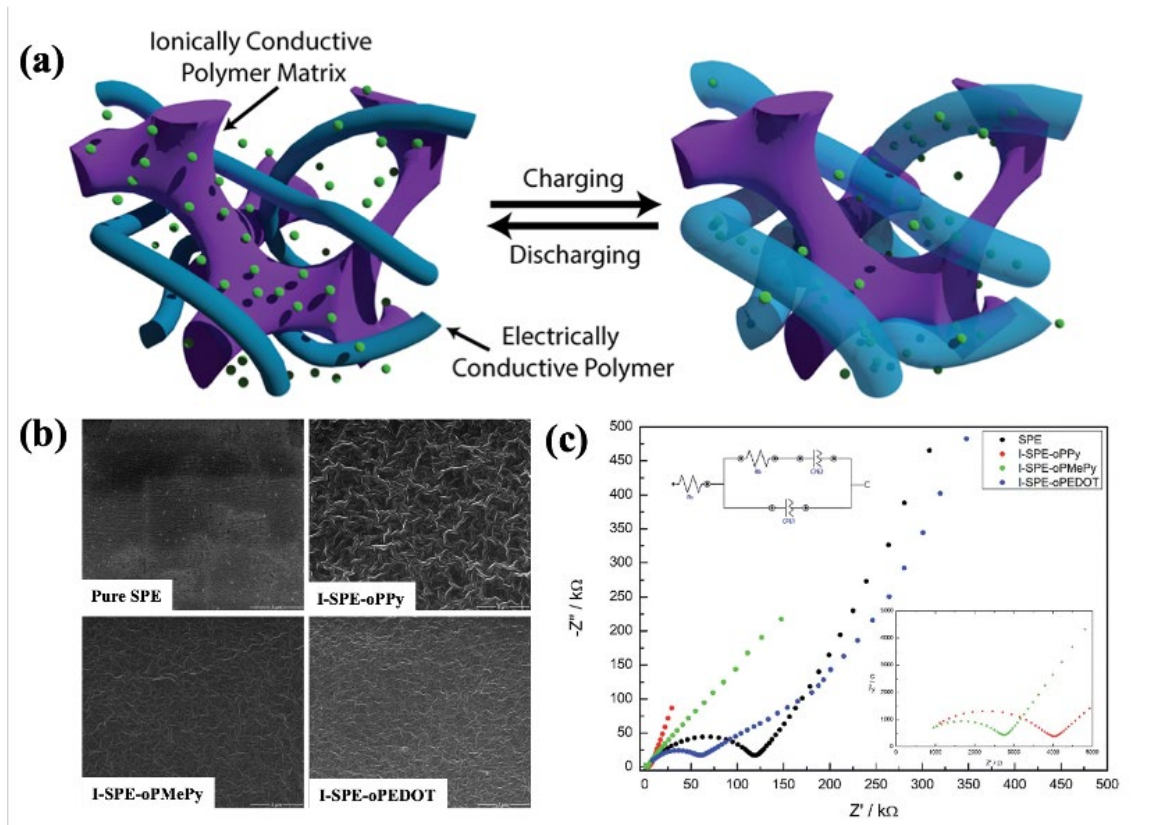
*Fig. 8. (a) Schematic illustration of semi-IPN PEDOT-PEO matrix. The cross-linked PEO matrix acts as a reservoir of electrolyte ions and locally constraints swelling of the PEDOT during charging-discharging cycling [77]. (b) The morphological images of pure SPE, I-SPE-oPPy, I-SPE-oPMePy, and I-SPE-oPEDOT. The pyrrole-derivatives I-SPE-oPPy (b) and I-SPE-oPMePy (c) display a uniform network without any depositions. Consequently, there is a link between the materials' structural analysis and their ionic conductivity [81].*

Leš et al. [83] in their work reported SPEs with the encapsulation of the CPs; poly(1H-pyrrole) (I-SPE-oPPy), poly(N-methyl pyrrole) (I-SPE-oPMePy), and poly(3,4-ethylene dioxythiophene) (I-SPE-oPEDOT). Each CPs was overoxidized subsequently by an electrochemical procedure. The morphology of the fabricated membranes is shown in Fig. 8 (b). The surface of the SPE polymer matrix pure SPE is quite dense with few inhomogeneous regions with pores (areas with brighter contrast). As a result, the LiTFSI is not uniformly distributed throughout the matrix. The polymer composites, on the other hand, construct a 3D structure surface despite having significantly thicker polymer chains. More space was observed between the individual polymer pieces. The I-SPE-PEDOT membrane exhibits some crystals attached to the polymer chains. A link between the dissociation of the Li salt in the polymer matrix and the ionic conductivity is dominant since the dissolution leads to a bigger number of free ions for Li transport [84]. Meanwhile, in Fig. 8 (c), the I-SPEs-oPMePy exhibited higher ionic conductivity at $7.00 \times 10^{-6}$ S/cm compared to pure SPE at $1.3 \times 10^{-7}$ S/cm. The calculated activation energies for Li-ion conduction display the effect of the generated IPNs, which lower the energy barrier for Li$^+$ conduction. Such materials could lead to a novel group of interpenetrated solid polymer electrolytes as separator materials based on mixed conductors. Moreover, the combination of a polyacrylate-matrix with cation-selective properties of overoxidized conducting polymers leads to 3D materials with higher ionic conductivity than SPEs and selective ion-exchange membrane properties with good stability by

facile fabrication. These kinds of synthesis methods appear to provide an alternative way of varying the properties and applications of CPs.

**Conclusions and Future Outlooks**

In the last decade, Li has emerged as a highly sought after resource. The rapid increase in demands for Li in lithium batteries (LiBs) for various applications such as electric vehicle batteries, energy storage grid systems, and personal electronics during the past few years have prompted concerns about the supply among the international community. Despite the resource reserve, the environmental sustainability of Li extraction, production and recovery is also critical. However, the current methods of Li extraction such as pyrometallurgy and hydrometallurgy are time-consuming and energy-intensive. In addition, these procedures are harmful to the environment because of the large amounts of chemicals used. Therefore, there is a lot of interest all around the world in refining and improving existing Li extraction in green methods. The electrochemical extraction method is seen as more environmentally benign due to manifold benefits/advantages, such as (1) produced less chemical waste production, (2) operation (OR could be operated) at room temperature, (3) does not require much energy to operate and, (4) it could yield Li up to 99% purity. This approach is capable of extracting Li from LIBs using a separator membrane, as explained in this article. The use of SPEs and CPEs-based membranes for Li harvesting provides many benefits, including (1) a low-cost operation, (2) high separation efficiency, (3) high selectivity, (4) high permeability. In addition, the separation process is less harmful to the environment. Sadly, membrane technologies have fallen victim to their own advantages. Membrane fouling, membrane longevity, and difficulties in scaling up operations are all drawbacks of membrane technologies. Poor physical and ionic conductivity of membranes are also some of the significant obstacles, necessitating more development in the future. Therefore, modification of the membranes and the addition of fillers are often used to scale up their ionic conductivity. The introduction of those fillers could decrease the polymer's crystallinity, which significantly improves ion migration and ionic conductivity. Li salts, ionic liquids, composite polymers, and interpenetrating network polymers are all examples of modified membranes. This includes focus on developing novel membranes with enhanced selectivity and stability, as well as optimizing existing procedures. For instance, using CPs integrated into the SPE membrane could tune a specific framework architecture and present promising avenues for developing continuous Li extraction in green recycling.

**Conflicts of Interest**

There are no conflicts of interest to declare.

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