리튬이온전지 폐기물로부터 가치 있는 리튬금속을 멤브레인 기반으로 회수

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Membrane Based Recovery of Valuable Lithium Metals from Lithium Ion Battery Waste

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요   약: 환경오염을 제어하기 위한 청정에너지에 대한 수요 증가는 빠르게 증가하고 있습니다. 리튬 이온 배터리와 같은 충전식 배터리는 청정에너지의 우수한 원천이지만 높은 수요와 공급 불일치로 인해 리튬 금속이 빠르게 고갈되고 있습니다. 배터리 폐기물에서 귀금속을 회수하는 것은 환경오염 제어와 함께 가능한 해결책 중 하나입니다. 멤브레인 기반 분리 방법은 폐기물에서 리튬을 회수할 수 있는 매우 성공적인 상업적 공정입니다. 이 작업은 최근에 보고된 다양한 방법을 다룰 것이며 검토 형식으로 작성될 것입니다.

Abstract: Growing demand on clean energy to control environmental pollution is growing rapidly. Rechargeable battery such as lithium ion battery is excellent source of clean energy but there is rapid depletion of lithium metal due to high demand and supply mismatch. Recovery of the precious metal from the battery waste is one of the possible solution along with the environmental pollution control. Membrane based separation method is highly successful commercial process available to recover lithium from the waste. This work will cover various methods reported recently and will be compiled in the form of a review.

Keywords: LIB, membrane, waste, AEM, dialysis

1. Introduction

With the rapid advancements of modern technologies, such as mobile gadgets and electric vehicles, the need for valuable lithium metals intensifies. One of the major sources of lithium is brine, which is obtained after the process of desalination of seawater[1]. However, as the demand for lithium increases faster than supply, novel approaches for recovery of lithium from wasted lithium ion batteries (LIBs) emerged. Thus, recycling of lithium from LIBs not only addresses the lack of resources but also provides a solution for the modern environmental issues related to pollution.

Lithium ion batteries are a key component of energy supply in current high-tech electronics. A crucial component of LIBs is the separator, which prevents internal short circuits and promotes the rapid lithium ion migration. Common separator materials such as poly-

ethylene (PE) and polypropylene (PP) experience limitations like poor thermal resistance and restriction of ion migration. Therefore, polyvinylidene fluoride (PVDF) based separators are now used in LIBs to enhance the thermal stability and wettability[2]. However, strong bonds between cathode materials, Al foil and binders like PVDF poses challenges for the recovery of lithium. It is crucial to perform pre-treatment, involving methods like physical separation or thermal treatment to ensure the total separation of cathode materials from the substrate, and to focus on the recovery of precious elements[3].

Electrochemical lithium recovery (ELR) system is one of the methods of lithium recovery, that promotes faster lithium extraction compared to conventional methods[4]. To address the challenge of recovering of lithium with low concentration, studies, involving ELR systems employing λ-MnO$_2$ electrodes, have been conducted[5,6]. The findings revealed that lithium recovery performance can be enhanced by reducing the size of λ-MnO$_2$ electrode particles to increase the electrode/electrolyte interface.

Developing more ecological alternative is crucial to employ these methods to real-life applications. Therefore, lithium manganese oxide (LiMn$_2$O$_4$) is used as a cathode material owing to its low cost, and sustainability[7,8]. In addition, the use of polyaniline (PANI) prevents the quick capacity loss caused by low reversibility of the chloride-capturing negative electrodes.

One more effective technique for ion selective separation with little effect on the environment is electrically switched ion exchange (ESIX)[9]. By adjusting the redox states of electroactive ion exchange films, ESIX technology precisely extracts ions from liquids. The ion-coordinated films based on montmorillonite (MMT) material presents the greatest potential among them due to the availability of plenty of exchangeable sites. Thus, PANI/MMT composite exhibits quick and precise ion separation[10].

A further difficulty in lithium recovery is coexisting ions, particularly magnesium, which has a lot of similarities with Li$^+$. Hybrid capacitive deionization (HCDI)

is one of the solutions to this problem. HCDI utilizes a cell with a lithium spinel-type sorbent cathode and an activated carbon anode coated with an anion exchange membrane (AEM), which inhibits re-sorption of co-ions during desorption[11,12]. To improve the efficiency of lithium recovery, there is a need to focus on electrode materials and cell designs. For example, the spinel layered heterostructure lithium-rich material (LSNCM) with high lithium extraction capacity, and nanocrystalline bismuth (ncBi) electrodes, that inhibits lithium ion exchange, are utilized to achieve this goal[13].

Overall, membrane-based recovery has shown promise in addressing these issues by providing efficient methods of recovering valuable lithium metals from lithium ion battery waste. In this review, numerous membrane based methods are reviewed and discussed in detail.

2. Recovery of Lithium

Diffusion dialysis (DD) is a membrane separation technique that operates based on the concentration gradient throughout the membrane[14]. Diffusion dialysis utilizes ion-exchange membranes (IEM), which are typically divided into two categories based on the way they operate as separation media: anion-exchange membranes (AEM), and cation-exchange membranes (CEM). In electrodialysis (ED) an electric field drives the migration of anions and cations through AEM and CEM, accordingly[15].

AEMs, which are employed in DD and ED, contain positively charged groups and attract anions more strongly due to electrostatic forces[16]. It allows anions to pass through the membrane while cations co-transport with the anions via the AEMs to preserve electrical neutrality. The membranes' electrostatic repulsion and steric hindrance make it easier for cations with shorter hydrated radii and smaller charges to pass through. Therefore, this mechanism facilitates the separation of cations.

Nanofiltration (NF) is a pressure-driven separation membrane technique that display separation character-
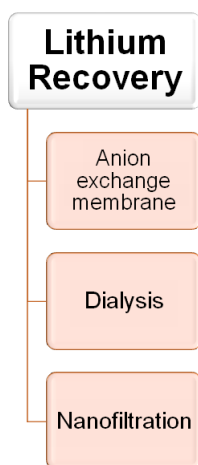
Fig. 1. Schematic representation of classification of the review.

istics between reverse osmosis and ultrafiltration membranes[17]. The combined impacts of size sieving and electrostatic exclusion primarily dictate the separation efficiency of NF membranes. These membranes offer potential for the selective removal of Mg^{2+} and $Li+$ since they can eliminate multivalent ions while allowing monovalent ions to move through. Fig. 1 represents the schematic representation of the classification of the review.

Repurposing discarded lithium batteries offers a valuable opportunity to recover precious lithium metals and benefit the environment[18]. Hierarchically structured membranes, particularly those utilizing modified metal-organic frameworks (IL-MOF), show promise for lithium recovery. Han *et al.* created the modified metal-organic framework (IL-MOF) by attaching MIL-101 (Cr) to its coordinate unsaturated site (CUS) using various ionic liquids (ILs). This is followed by developing a flexible hydrophilic thin film nanocomposite (TFN) composed on polyvinylidene fluoride (PVDF) using IL-MOF nanoparticles. Thorough characterizations of the TFN membrane based on IL-MOF are conducted, followed by empirical lithium recovery testing using a simulated leaching solution of lithium-ion batteries (LIBs). Results show that IL-MOF nanoparticles significantly enhance lithium selectivity, with a fourfold increase for $S_{Li^+,Mn^{2+}}$ (from 1.73 to 8.91), $S_{Li^+,Co^{2+}}$ (from 1.75 to 9.94) and $S_{Li^+,Ni^{2+}}$ (from 1.69 to 10.09) compared to the original membrane. Over five filtration cycles, 98.9% of lithium is recovered while maintaining membrane integrity.

Kang *et al.* proposed a method of green recycling of the solvent in manufacturing of LIBs, which utilizes self-cross-linked nanocomposite membranes[19] (Figs. 2, 3).

Composite membranes made of polyvinyl alcohol (PVA) and carbon nanotubes (CNTs) were made by employing carboxylated CNTs as a substitute to cross-linkers. Cross-linking between CNTs and PVA chains has been verified by molecular modeling, which projected its stability in water. The idea was supported by the composite membranes, which included probe sonicated CNTs and demonstrated resilience against water dissolving at 80°C with 1.5 wt% CNT loading. High separation efficiency was demonstrated by membranes for the recycling of 1-methyl-2-pyrrolidon (NMP) in the production of LIBs. Consistent flux and water/NMP selectivity of 0.06 kg/m²·h and 3500 were shown after long-term usage of membranes using 1.5 wt% CNTs, thereby improved sustainability in the LIB manufacturing sector.

By employing sustainable hydrolysis polymerization, Qu *et al.* developed a lithium ion-imprinted membrane (LIIM)[20]. LIIM demonstrated fast Li^+ adsorption in accordance with a pseudo-second-order kinetic model, where chemical adsorption is connected to activation energy. LIIM proved spontaneous exothermic Li^+ adsorption by thermodynamic analysis. In the simulated leaching solution (SLS), LIIM exhibited a significant affinity for lithium. In addition, LLIM demonstrated high selective separation factors of Li^+ over Fe^{2+}, Cu^{2+}, Ca^{2+}, Zn^{2+}, Mn^{2+}, K^+, and Na^+ being 10.93, 19.30, 22.66, 23.49, 26.14, 27.57, and 39.00, respectively, indicating satisfactory performance. The findings demonstrate that LIIM is a highly selective, environmentally friendly adsorbent with extensive potential for recycling wasted LIBs.

With the incorporation of WS_2 nanofillers (M7) in polymeric membranes, Santoro *et al.* explored the us-
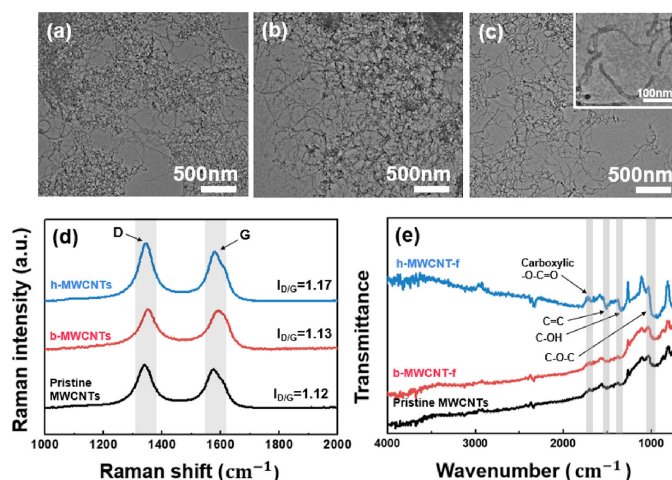
Fig. 2. Physicochemical properties of the CNT samples: TEM images of MWCNTs of (a) pristine MWCNTs, (b) b-MWCNTs, and (c) h-MWCNTs; (d) Raman spectra of pristine MWCNTs, b-MWCNTs, and h-MWCNTs; and (e) ATR-FTIR spectra of pristine MWCNTs, b-MWCNT-f, and h-MWCNT-f (Reproduced with permission from Kang *et al.*[15], Copyright 2022, American Chemical Society).
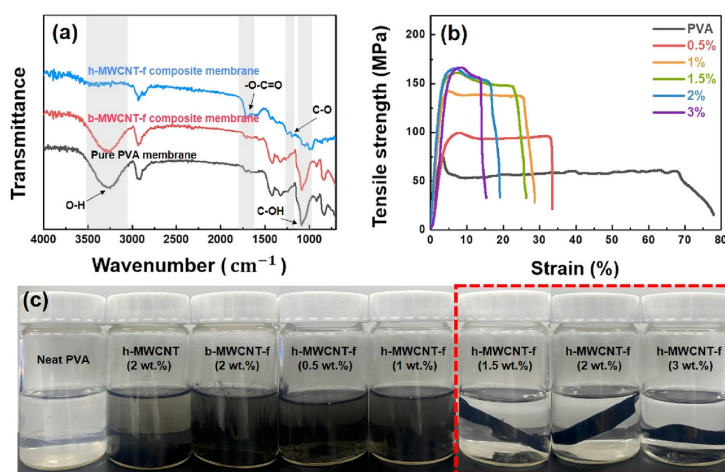

Fig. 3. Physicochemical properties of PVA/CNT composite membranes: (a) ATR-FTIR spectra of the neat PVA, b-MWCNT composite, and h-MWCNT-f composite membranes; (b) tensile strength of the neat PVA and h-MWCNT-f composite membranes (0.5~3 wt%); and (c) stability test results of the neat PVA, h-MWCNT (2 wt%), b-MWCNT-f (2 wt%), and h-MWCNT-f composite membranes (0.5~3 wt%) (Reproduced with permission from Kang *et al.*[15], Copyright 2022, American Chemical Society).

age of exciton-based light-to-heat conversion to induce photothermal membrane crystallization, enabling optimal lithium extraction from brines rich in lithium[21]. The water evaporation flux in PVDF-WS$_2$ nanocomposites is increased by 364% as photothermal effects are induced. Under the condition of supersaturation, this results in the heterogeneous nucleation as well as crystallization of LiCl salt, achieving an 8.4% recovery, which outperformed unloaded mem-

brane (M0). Functional inks based on van der Waals semiconductor nanosheets are gaining popularity since now they are being used in the recovery of raw materials and crystallization. These discoveries present a potential of environmentally friendly extraction of lithium for technologies of renewable energy.

2.1. Anion exchange membrane

In response to the rising demand for lithium, a novel

electrochemical approach has been developed for the recovery of lithium. It follows the basic concepts of lithium-ion batteries by using silver as a negative electrode and a lithium-capturing electrode[22]. Kim *et al.* suggested zinc as a cost-effective alternative to silver due to its affordability, high capacity, and stable redox potential. It resulted in an efficient extraction of lithium, achieving an energy consumption of 6.3 Wh mol^{-1} of recovered lithium through a $LiMn_2O_4$–zinc (LMO-Zn) battery system. Zinc electrode oxidation and reduction were reversible without any electrode-potential fluctuations when a high concentration of $ZnCl_2$ was maintained in a small compartment. Through 100 rounds of charging and discharging, the LMO-Zn system's stability was demonstrated, maintaining 73% of its original capacity without zinc waste.

A unique method for recycling $LiCoO_2$ from spent LIBs is put forward, where Liu *et al.* used a Zn-air desalination battery (ZADB)-based three-channel ion recovery device that can extract Li^+ and Co^{2+} from the recovered solution and provide energy at the same time[23]. Ion mobility is facilitated by the device's anion exchange (AEM) and cation exchange membranes (CEM), which divide three chambers. Finite element simulation (FES) reveals the migration of Li^+ and Co^{2+} to the cathode chamber, with the generation of OH^-. Meanwhile, SO_4^{2-} travels to the chamber of the Zn foil anode and combines with Zn^{2+} ions. The highest recovery efficiency is achieved under specific conditions: using a 0.2 mol L^{-1} recovered solution at 0.2 mA cm^{-2} discharge current density for 24 hours. The device exhibits an output energy density of 102.5 Wh Kg^{-1} and an average recovery rate of 0.275 mg min^{-1}. It also has outstanding recycling stability and long-term cycle performance.

2.2. Dialysis

Wu *et al.* introduced an integrated method of diffusion dialysis (DD) and electrodialysis (ED) to address the environmental concerns caused by wastewater generated during lithium battery recycling, containing acids and heavy metals[24] (Fig. 4).
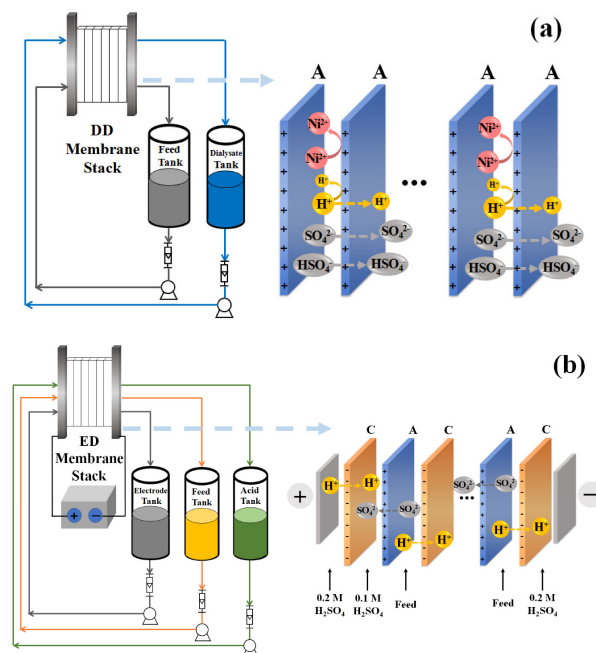

Fig. 4. The diagram of (a) the DD set-up and membrane stack; (b) the ED set-up and membrane stack (Reproduced with permission from Wu *et al.*[20], Copyright 2023, American Chemical Society).

The main objective of this method was to extract Ni^{2+} and H_2SO_4 from the wastewater. The process starts with acid and heavy metals being separated using the DD method. High recovery rate of 75.96% for acid and rejection rate of 97.31% for Ni^{2+} were achieved in the DD process under specific conditions: a 300 L/h flow rate and a 1:1 W/A flow rate ratio. Next, a two-stage ED setup is utilized in the ED process to concentrate the recovered acid from 43.1 g/L to 150.2 g/L H_2SO_4, making it suitable for application in the battery recycling. With potential for industrial use, this integrated method presents a viable approach to battery wastewater treatment.

Utilizing hydrometallurgy integrated with membrane separation technology offers novel approaches for the recovery of lithium from spent LIBs[25]. Xie *et al.* used simulated spent battery leachate to develop and test positively charged poly(ionic liquid) membranes with preserved liquid crystal mesophases for monatomic ion separation. The findings demonstrated that bicontinuous cubic liquid crystal membranes exhibited
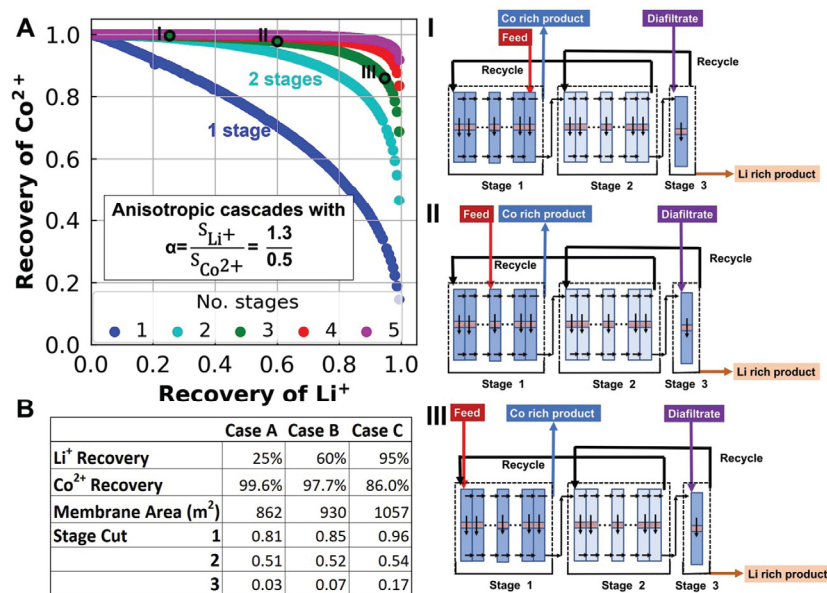
Fig. 5. Pareto set (A) is obtained by solving the superstructure optimization problem for the design of isotropic diafiltration cascades utilizing a membrane with selectivity $\alpha = 2.6$. Each point in the Pareto set, which represents a solution to the optimization problem and corresponds to a complete physical design for an isotropic cascade, was obtained by maximizing the Co^{2+} recovery while constraining the Li^+ recovery to a specific value. Three representative three-stage configurations that highlight the heterogeneity of physical designs that can emerge with respect to recovery, membrane area, stage cut, and feed injection strategy are described in panel B and panels I–III. Design (I) at 25% Li^+ recovery has all the feed entering the first stage of the cascade. Design (II) at 60% Li^+ recovery has a split feed. Design (III) at 95% Li^+ recovery has the feed entering the last stage of the cascade. These three designs reveal a nonintuitive relationship between the targeted Li^+ recovery and the optimal location of the feed stream. Calculating the 151 solutions for the three-stage Pareto set, from which the three illustrations are taken, took 1 min and 37 s, for an average time of 0.64 s to compute each optimal design (Reproduced with permission from Wamble *et al.*, 23, Copyright 2022, American Chemical Society).

an accurate ion separation, particularly for enriching lithium ions. Membranes with this bicontinuous cubic structure demonstrated three times greater ion flux than amorphous membranes and improved ion selectivity as compared to other structural types. The Li^+/Ni^{2+} selectivity reached 7.40, surpassing that of commercial (5.71) and hexagonal liquid crystalline membranes (2.59).

2.3. Nanofiltration

Kumar *et al.* proposed a unique membrane-integrated hybrid system that aims to recycle precious metals from acidic leach liquor that comes from wasted LIBs[26]. The process involves pretreating the leach liquor with alkaline solutions (NaOH and NH_4OH) to adjust its pH to 5.53, followed by ultrafiltration to remove Fe and Al and reduce turbidity (~1.6 NTU).

Subsequently, Ni^{2+}, Co^{2+}, and Mn^{2+} ions were effectively separated by nanofiltration membrane (VNF2), with rejection values of 92.5%, 94.6%, and 95.8%, respectively. When bivalent ions are fractionated into monovalent ions, the concentrations of Ni^{2+}, Co^{2+}, and Mn^{2+} ions increase from 0.74, 0.52, and 0.63 g/L to 6.14, 4.59, and 5.62 g/L, respectively, preserving 90% feed solution recovery. Lastly, there is a crystallization of Li^+ in the nanofiltrate to Li_2CO_3, with a recovery percentage of 88.2% and a purity of 99.5 wt% attained at 70 C operating temperature with 4M of K_2CO_3.

The demand for materials like cobalt, lithium, nickel, and manganese from wasted LIBs drives innovative recycling techniques to address electronic waste issues [27] (Fig. 5).

The intricate leaching and extraction methods to recover Co^+ and Li^+ rely on high temperatures and or-

ganic solvents. Therefore, Wamble *et al.* proposed a method that uses harmless continuous membrane cascades. In order to build diafiltration cascades, that boost material purity and recovery, a superstructure optimization model is devised. This model predicts the best trade-offs between lithium and cobalt recovery and purity, enabling immediate evaluations of membrane materials. In a two-stage cascade arrangement, a nanofiltration membrane that has moderate selectivity may recover 95% Li^+ and 99% Co^{2+} with a purity of 93 and 99.5 wt%. An investigation of more than a thousand Pareto best designs reveals six design heuristics for carrying out binary separations employing staged diafiltration cascades. Additionally, this study provides new possibilities for membrane-based LIB recycling by analyzing membrane materials in tailored diafiltration procedures.

3. Conclusion

The recovery of valuable lithium metals from lithium-ion battery waste has progressed significantly due to the optimization of membrane separation methods. Advanced techniques and materials, such as hierarchically structured membranes using modified metal-organic frameworks (IL-MOF) and innovative nanocomposite membranes, have shown promising results in enhancing lithium recovery efficiency. The use of IL-MOF nanoparticles significantly improves lithium selectivity and recovery rates, while composite membranes made from polyvinyl alcohol (PVA) and carbon nanotubes (CNTs) exhibit high separation efficiency and stability. Innovative materials including WS_2 nanofillers improve polymeric membranes' separation capabilities and structural integrity, promoting environmentally friendly recycling methods. Moreover, the method for recycling $LiCoO_2$ from spent LIBs using a Zn-air desalination battery-based device proposes the integration of energy generation and lithium-cobalt recovery, showcasing the multifunctionality of modern recycling technologies. Lastly, the method of using

continuous membrane cascades for lithium and cobalt recovery presents an optimized and safe alternative to traditional high-temperature and solvent-dependent methods. Their superstructure optimization model predicts the best trade-offs between recovery and purity, providing valuable insights for future membrane-based LIB recycling technologies.

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