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Recycling lithium-ion batteries from electric vehicles using environmentally friendly chemical and biological methods in the trend of developing a circular economy

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ABSTRACT
Amid increasing awareness of environmental pollution and the necessity for developing a circular economy, recycling lithium-ion batteries has become an undeniable means to establish a sustainable and eco-friendly electric vehicle operation system. This article focuses on introducing and analyzing chemical methods for recycling lithium-ion batteries, with particular emphasis on green chemical and biological approaches. By applying these methods, we can recover valuable materials such as lithium, cobalt, and nickel from used batteries, creating a closed-loop material cycle and reducing electronic waste. Furthermore, recycling lithium-ion batteries using these green methods also brings economic benefits by creating new business opportunities and reducing production costs. Thus, this article highlights the role and potential of green chemical and biological methods in recycling lithium-ion batteries within the context of developing a circular economy.

Keywords: *electric vehicles; lithium-ion batteries (LIBs), chemistry, biology, circular economy.*

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I. INTRODUCTION

In the 21st century, transitioning to clean and sustainable transportation has become a top priority in the world's efforts to minimize negative environmental impacts and create a pathway for sustainable development. Among these modes of transportation, electric vehicles have emerged as a potential solution, offering advantages in reducing emissions and fossil fuel consumption. However, a challenge for electric vehicle technology is the management and recycling of lithium-ion batteries, a key component in the vehicle's energy system.

Lithium-ion batteries play a crucial role in the operation of electric vehicles, providing the energy source to power electric motors and other electronic systems. However, the issue of recycling lithium-ion batteries has become a complex challenge for the electric vehicle industry, as millions of these batteries need replacement annually due to capacity loss and decreased performance over time.

In this context, the need for recycling lithium-ion batteries is not only a technical issue but also an opportunity to promote the development of a circular economy. Green chemical methods are becoming an integral part of this recycling process, offering not only environmental benefits but also creating economic value from recycled resources.

This article focuses on examining modern green chemical methods applied to recycle lithium-ion batteries in electric vehicles. We will evaluate the progress in this field, from the recycling processes to practical applications in the industry, as well as the challenges and opportunities that these methods present.

II. Overview of the Current Situation of Lithium-Ion Battery Recycling

Lithium-ion battery recycling is an integral part of sustainable development in the electric vehicle industry. Currently, recycling methods mainly rely on physical techniques but are facing pressure from increasing demands for efficiency and environmental considerations.

- Reuse: One of the most common methods is reusing functional components of lithium-ion batteries, although this often encounters limitations in efficiency. Battery cells can be disassembled and reused in smaller applications or as backup power sources, but this does not guarantee high efficiency and can lead to rapid capacity and performance loss.
- Material Recycling: This method involves separating battery components and recycling them into input materials for new battery production. However, this process may face challenges in efficiently and safely separating components without causing environmental pollution.

- Energy Recovery: A portion of lithium-ion batteries cannot be reused, and this recycling method typically involves incineration or high-temperature processing to extract energy. However, this process may pose safety and environmental pollution issues.

Although these methods have been applied, they still face many limitations. The use of green chemical methods is becoming a potential direction to address these issues but requires significant investment in research and development of new technologies. This presents a major challenge but also provides significant opportunities for the advancement of the electric vehicle industry and the recycling industry as a whole.

III. Opportunities and Challenges for LIBs Recycling

***Opportunities:**

- Recycled Resources: LIBs contain rare materials such as lithium, cobalt, nickel, and aluminum. Battery recycling helps reuse these resources, reducing pressure on supply sources and saving natural resources.
- Environmental Pollution Reduction: The process of producing new batteries generates large amounts of emissions and environmental pollution. Recycling reduces electronic waste and pollution from new battery production.
- Economic Recycling: LIBs recycling can create economic opportunities by recovering valuable materials and producing ancillary products for other industries.
- Technology Development: Research and development of LIBs recycling methods also mean the development of green, sustainable technologies, creating opportunities for businesses and researchers.

***Challenges:**

- Technical Complexity: The LIBs recycling process is technically complex due to the combination of various materials and chemicals. This requires significant investment in research and development of recycling technologies.
- Environmental Safety: The recycling process must be carried out safely to avoid environmental pollution and ensure the health of workers in the industry.
- Cost: Currently, LIBs recycling may still be more expensive than producing new batteries due to the cost and development of recycling technologies. Risk Management: LIBs recycling requires careful risk management to ensure safety for both workers and the environment, including handling hazardous chemicals and fire prevention.

In summary, lithium-ion battery recycling offers many opportunities for environmental and economic sustainability but also faces challenges in technical, safety, and cost aspects. To leverage these opportunities and overcome challenges, collaboration among governments, businesses, and research organizations is needed.

IV. Environmentally friendly methods LIB battery recycling

The explosive growth of electric vehicles (EVs) has led to a threefold increase in lithium prices and a fourfold increase in cobalt prices from 2016 to 2018. To reduce production costs and product prices, as well as to protect the environment, there is a demand for processes that recover and reuse these valuable materials. Generally, recycling relies on first-generation recovery technologies, which involve physical processing to obtain various raw materials, followed by hydrometallurgical processes (filtration and extraction) to separate metals.

In fact, lithium-ion batteries (LIBs) typically use a positive electrode made of lithium metal oxide, which commonly includes lithium iron phosphate (LFP), lithium nickel cobalt manganese oxide (NMC), lithium nickel cobalt aluminum oxide (NCA), lithium manganese oxide (LMO), or lithium titanate oxide (LTO). First-generation LIBs, primarily used in handheld electronic devices, utilize lithium cobalt oxide (LCO).

Battery cells are assembled into modules, and modules are further assembled into battery packs. The voltage from the battery power source provides current to electric vehicle (EV) motors, which can reach up to 300 V or even exceed 600 V.

By weight percentage (g material/g battery), lithium-ion batteries typically consist of approximately: 7% Co, 7% Li (represented as equivalent to lithium carbonate, 1 g lithium ¼ 5.17 g LCE), 4% Ni, 5% Mn, 10% Cu, 15% Al, 16% graphite, and 36% other materials [10].

Solid-state electrolysis produces mainly stable components (such as Li_2CO_3) and highly stable compounds (polymers, $ROCO_2Li$, $(CH_2OCO_2Li)_2$, and ROLi easily decomposed upon heating to >90 ^oC, releasing flammable gas and oxygen), which gradually settle on the anode surface to form a passive film. This film layer limits electrochemical reactions by preventing LIB from accessing lead sites, thereby increasing internal ohmic resistance.

A typical EV lithium-ion battery pack has a useful lifespan of 200,000–250,000 km on the first use, although rapid charging at >50 kW is increasingly applied, reducing battery life due to rapid degradation.

When an EV battery pack loses 20% (15% for certain electric vehicle models) of its initial capacity, it becomes unsuitable for propulsion because the reduced battery capacity affects acceleration, range, and vehicle regeneration capability.

According to a comprehensive analysis conducted by Melin in 2017, by 2025, approximately 75% of used electric vehicle batteries will be reused in second-life solutions for several years after they are no longer used in vehicles. They will then be recycled to recover all valuable components.

*A new green chemical technology for recycling lithium-ion batteries from electric vehicles utilizes citric acid (H3Cit) and H_2O_2 solution to recover metals with high efficiency (98% Co and 99% Li). Used batteries are manually disassembled to recover Al and Cu foils in metal form and separator devices, which are then directly recycled. The cathode waste material is crushed and calcined at 700˚C for 2 hours before further processing. The H_2O_2 solution is used as a clean reducing agent in the metal filtration process, with both metal ions and citric acid simultaneously recovered by selective precipitation. Co and Li ions are processed using oxalic acid and phosphoric acid for recovery. This process allows for the recovery of nearly 99% Co and 93% Li in the form of $CoC_2O_{42}H_2O$ and Li_3PO_4, respectively, while citric acid recycling capabilities are similar to fresh acid. Additionally, this process also allows for direct recycling of the negative electrode to produce new LFP negative electrodes for new batteries.

***Biotechnological Approaches:** Biotechnological methods utilize microorganisms or enzymes to selectively extract metals from battery materials. This emerging field, known as bioleaching or biomining, offers the potential for environmentally friendly and energy-efficient metal recovery processes. However, further research is needed to optimize these techniques for lithium-ion battery recycling.

The biological leaching process for extracting metals from LIBs is an environmentally friendly and cost-saving method with lower energy requirements and greenhouse gas emissions compared to traditional pyrometallurgical and hydrometallurgical processes (Krebs et al., 1997; Hoque & Philip, 2011; Kaksonen et al., 2018). However, challenges in the biological leaching process are directly related to hazardous components such as metals, binders, electrolytes, and secondary reactions. A diverse group of microorganisms involved in metal bioleaching includes lithotrophic bacteria, chemoautotrophic bacteria, and fungi (Hedrich et al., 2011; Johnson, 2014; Quatrini & Johnson, 2019). Originally developed for ore processing, bioleaching technology is now being applied to LIB recycling. Over the past 30–40 years, 20 bioleaching plants for metal extraction from ores have been constructed. Four groups of microorganisms can be used for LIB recycling based on their nutritional requirements (autotrophic, heterotrophic, and mixotrophic) (Bosecker, 1997; Johnson & Roberto, 1997; Mishra & Rhee, 2014; Jafari et al., 2018). According to metabolic temperature, these bacteria are classified as mesophiles (below 40°C), thermophiles (45°C–70°C), and hyperthermophiles (above 70°C) (Duarte et al., 1993; Norris, 1997; Zhao & Wang, 2019). Bacteria utilize inorganic and carbon-based materials as their energy sources, termed lithotrophic and heterotrophic organisms, respectively; however, some heterotrophic organisms can metabolize energy under anaerobic conditions without oxygen. These microorganisms can be used individually and in consortia to assess the overall efficiency of the bioleaching process (Is¸ıldar et al., 2017; Yu et al., 2020).

The bioleaching process is sustainable, environmentally friendly, cost-effective, and energy-efficient with low greenhouse gas emissions for metal recovery from LIBs. Microorganisms used in the bioleaching process produce bio-sulfuric or organic acids. Bacteria oxidize iron ions to ferric ions, which are then used as reducing agents to convert the oxidation state of metal ions. These bacteria can dissolve metals from LIBs through acid dissolution and redox reactions. Fungi produce organic acids that can dissolve metal ions with the assistance of oxidizing agents such as H_2O_2 through complexing reactions. However, the bioleaching method has limitations for extracting metals from LIBs on a large scale due to significant challenges such as long incubation times, slow process kinetics, low solid-liquid ratios (paper pulp density), and metal toxicity. The success of the bioleaching process depends on the production of transformation compounds, microbial tolerance to hazardous metals, bacterial inoculation with low-cost nutrients, and enhanced process kinetics.

V. Efficiency in both economic and environmental aspects

In a recent patent, seven main components (cobalt, lithium, copper, lead, nickel, aluminum, and manganese) were reported to account for >90% of the economic value of used lithium-ion batteries: Co (39%) and Li (16%, equivalent to LCE), followed by Cu (12%), Pb (10%), Ni (9%), Al (5%), and Mn (2%).

Economic efficiency:

1. Resource savings: Recycling lithium-ion batteries helps save rare resources such as lithium, cobalt, and nickel. Reusing these materials reduces the cost of new production and maintains product prices more stable.
2. Reduction in waste disposal costs: Recycling reduces the amount of hazardous electronic waste introduced into the environment, lowering the costs of processing and disposal. Avoiding landfilling or incinerating waste also reduces expenses and negative environmental impacts.

3. Creation of secondary resource streams: Recycling processes can generate secondary resource streams, such as recycled metals, which can be resold or used in other applications, providing additional income for recyclers.

Environmental efficiency:

1. Waste reduction: Recycling lithium-ion batteries reduces the amount of hazardous electronic waste released into the environment, mitigating negative impacts on soil, water, and air.
2. Reduction in air pollution: Reusing recycled materials helps reduce the need for strategic mining and extraction processes, lowering energy consumption and associated emissions.
3. Protection of water resources: Recycling processes cause less water pollution compared to new material extraction, reducing the risk of groundwater contamination and water environment.
4. Greenhouse gas reduction: Reusing recycled materials helps reduce the need for mining and new production, thereby lowering greenhouse gas emissions generated during manufacturing.

In summary, recycling lithium-ion batteries not only brings economic benefits but also has positive environmental impacts, reducing waste, saving resources, and minimizing environmental pollution. This promotes sustainable development in the battery industry and supports a greener economy.

The economic (and environmental) benefits of electric vehicles are significant and noteworthy. According to the 2017 report cited above, lithium recycling is expected to account for 9% of the total lithium battery supply by 2025 (specifically, 5,800 tons of recycled lithium, or 30,000 tons of LCE), with cobalt accounting for nearly 20% of the total supply. Furthermore, over 66% of lithium-ion batteries are recycled in China.

Overall, these two methods often provide high recycling efficiency and have less environmental impact compared to other recycling methods, but careful implementation is needed to ensure safety and maximum effectiveness.

VI. CONCLUSION

In the context of the circular economy becoming an important trend, leveraging green and biological chemistry methods to recycle electric vehicle lithium-ion batteries plays a crucial role in ensuring the sustainable development of the battery industry and supporting a greener economy. These methods not only help reduce waste, conserve rare resources, and decrease environmental pollution but also bring economic benefits by generating supplementary raw materials and reducing production costs. The combination of green and biological chemistry methods not only establishes an efficient recycling process but also opens doors for innovation and development in the automotive and battery industries. This highlights the importance of advancing research and implementing advanced and sustainable recycling methods to build a future where we can take pride in economic development and environmental protection. However, these studies need to continue evolving in the future to align with the global economy.

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