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## Processes and technologies for the recycling and recovery of spent lithium-ion batteries

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## ABSTRACT

LiBs pose a very specific threat, given that they contain a high percentage of dangerous heavy metals. From the 4000 t of used lithium-ion batteries collected in 2005, 1100 t of heavy metals and more than 200 t of toxic electrolytes were generated. This is why a lot of attention has been paid to the development of the technology necessary to recover and recycle LiBs in order not only to protect the environment but also to conserve resources. The recovery of major spent cell components is beneficial both in terms of environmental protection and also for the provision of raw materials. The authors of this article carried out a state of the art on the technologies used in the recycling and regeneration of industrial lithium-ion batteries. The main objective of such technologies is to enable the recycling of valuable elements present in the batteries, such as cobalt, nickel and copper, in a way which is both profitable and environmentally friendly. All the technologies used in the manufacture of lithium-ion batteries are constantly changing makes subsequent changes to the research into recycling and recovery technologies necessary. This does not mean merely finding ways to recover the precious metals, but also to recover other materials which may harm the environment, in order to dispose of them appropriately. The discussion of this research clearly reflects that:

- There are very few studies on the recovery of metals such as graphite, the electrolyte in spent LiBs, and it is our belief that more research is needed in this area.
- The research into the application of microorganisms in the used lithium batteries is few and far between.
- It is important to find ways to recover the precious metals and to recover other materials which may harm the environment, in order to dispose of them appropriately.

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## 1. Introduction

The purpose of this paper is to review the current status of the recycling and recovering technologies of spent lithium-ion batteries. It introduces the structure and components of the lithium-ion batteries and summarizes all kinds of recycling and recovery processes from spent lithium-ion. Also, the problems and prospects arising from the studies of their recycling technologies have been put forward.

Lithium batteries (LiBs) are generally composed of a cathode, an anode, an organic electrolyte and a separator. Lamination of the cathode, anode and separator, achieved through compression, makes electrical contact possible. The anode is a copper plate coated with a mixture of graphite, a conductor, binder polyvinylidene fluoride (PVDF) and additives such as  $\text{LiPF}_6$  [1].

Similarly, the cathode is an aluminum plate coated with a mixture of active cathode material, an electric conductor, a PVDF binder, and additives.  $\text{LiCoO}_2$  is commonly used as the active cathode material for almost all LiBs on the market [1]. Reference names for Li-ion batteries are shown in Table 1.

Advantages of LiBs such as their lightweight components, inflated energy capacity, high voltage per cell, favorable discharge resistance, ability to work through a large number of regeneration cycles, and a wide range of temperatures, alongside the fact that they are less harmful to the environment than other batteries, have meant that, since their introduction onto the market in 1991 [3], LiBs have been widely used in mobile electronic applications like PCs, video cameras, and mobile telephones, and later as energy storing devices in electric vehicles and stationary storage of renewable energies such a solar and wind. The success of lithium ion technology for the latter applications will depend largely on the cost, safety, cycle life, energy, and power, which are in turn controlled by the component materials used. Accordingly, several authors provide reviews focusing on the challenges and prospects associated with the electrode materials [4–7].

Table 2 provides an overview of the major class of lithium insertion electrode materials. Each system in Table 2 has its own advantages and disadvantages, which often dictate their application areas.

Global consumption of LiBs between the years 2000 and 2004 stood at around 500–700 million cells, whilst, according to the

International Telecommunications Union (ITU), the number of mobile telephone users exceeded 6.8 billion in 2013.

LiBs pose a very specific threat, given that they contain a high percentage of dangerous heavy metals. From the 4000 t of used lithium-ion batteries collected in 2005, 1100 t of heavy metals and more than 200 t of toxic electrolytes were generated [8,9,40].

Some of the components of these batteries are difficult to break down; meaning that discarding them after their end-of-life into municipal waste landfill sights may pollute the soil and underground water, while their incineration contaminates the air by releasing toxic gases [10].

Dorella and Mansur [11] ascertained the metal content of LiBs and found that valuable metals such as aluminum, cobalt, lead and lithium were the main components to be separated. They emphasized that irresponsible disposal of spent LiBs will result in environmental pollution.

This is why a lot of attention has been paid to the development of the technology necessary to recover and recycle LiBs in order not only to protect the environment but also to conserve resources. The recovery of major spent cell components is beneficial both in terms of environmental protection and also for the provision of raw materials [12–16].

## 2. Selection of research studies

This paper systematically reviews recent research on technologies used in the recycling and regeneration of industrial lithium-ion batteries. The main objective of such technologies is to enable the recycling of valuable elements present in the batteries, such as cobalt, nickel and copper, in a way which is both profitable and environmentally friendly. According to Dewulf et al. [17], the use of recycled cobalt and nickel in the production of the active cathode material present in LiBs leads to the following savings: 51.3% in natural resources, 45.3% in fossil fuels, and 57.2% in nuclear energy demand.

The methodology used for this systematic review is described in [18,19], and consists of the following steps:

**Table 1**  
Reference names for Li-ion batteries [2].

Chemical name	Material	Short form	Characteristics
Lithium Cobalt Oxide <sup>a</sup>	$\text{LiCoO}_2$ (60% Co)	Li-cobalt	Its high capacity makes it ideal for mobile phones, laptops and cameras.
Lithium Manganese Oxide <sup>a</sup>	$\text{LiMn}_2\text{O}_4$	Li-manganese, or spinel	This is the safest kind of battery, with a lower capacity than Li-cobalt but a high specific power and long life. Used in power tools, e-bikes. EV, medical, hobbyist.
Lithium Iron Phosphate <sup>a</sup>	$\text{LiFePO}_4$	Li-phosphate	
Lithium Nickel Manganese Cobalt Oxide <sup>a</sup>	$\text{LiNiMnCoO}_2$ (10–20% Co)	NMC	
Lithium Nickel Cobalt Aluminum Oxide <sup>a</sup>	$\text{LiNiCoAlO}_2$ (9% Co)	NCA	
Lithium Titanate <sup>b</sup>	$\text{Li}_4\text{Ti}_5\text{O}_{12}$	Li-titanate	Gaining importance in electric powertrain and grid storage.

<sup>a</sup> Cathode material.

<sup>b</sup> Anode material.

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