

# Recycling of lithium ion cells and batteries

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## Abstract

A new process for recycling lithium ion cells and batteries is described. In order to gain maximum value, the process aims to recover every component from the cell. In contrast with existing recycling processes, the AEA Technology process operates at ambient temperatures. There are three main stages; electrolyte extraction, electrode dissolution, and cobalt reduction. The technology is currently in development, with a demonstration unit linking the process stages together in active operation. Based on the projected quantities of lithium ion batteries available for recycling in the next few years, there is a significant market opportunity for a successful technology. © 2001 Elsevier Science B.V. All rights reserved.

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## 1. Introduction

Lithium ion cells and batteries will form an increasing proportion of the battery waste stream, as products using them reach the end of their practical life. It is therefore important that an optimum recycling process is available for this battery chemistry. This process should be safe, economic, and recover as much of the battery material as possible. The absence of metallic lithium gives greater flexibility in cell disassembly, and subsequent processing.

## 2. Contents of a lithium ion cell

Lithium ion cells contain spirally wound electrodes, insulated from each other by a porous separator material. The cathode (positive) electrode contains lithium cobalt oxide, carbon, and PVDF binder, coated on aluminium foil. The anode (negative) electrode contains carbon and PVDF binder, coated on copper foil. A liquid electrolyte is dispersed throughout the pores of the electrodes and separator. This is normally a solution of lithium hexafluorophosphate in two or more organic carbonates. The mass and approximate value for the main components in a standard ICR18650 cell are listed in Table 1.

From the table, it is clear that the cobalt is the most valuable component. However, the second most valuable component is the electrolyte. Therefore, any process seeking

to extract the maximum value from the cell will need to recover the electrolyte. This imposes process limitations, because the lithium salt is hydrolysed by water, and thermally unstable above about 80°C.

## 3. Existing lithium ion recycling processes

There are two established recycling processes for lithium ion cells and batteries, using extremes of temperature. The Toxco process [1] is designed for all types of lithium containing waste. The material is cooled in liquid nitrogen, before being mechanically shredded and mixed with water. The lithium reacts to produce hydrogen, which burns off above the reaction liquid. The main product is lithium hydroxide, but other components are “targeted where appropriate” (i.e. cobalt). The Sony process [2] uses a much higher temperature; the cells are incinerated. The metallic waste is recovered for processing to recover the cobalt, using standard hydro-metallurgical techniques. The organic components, lithium, and fluoride are lost, though a scrubbing system on the incinerator avoids emission problems. Larger cell sizes have to be punctured before they are introduced into the incinerator.

## 4. AEA technology recycling process

Having reviewed the existing processes, there is scope for an alternative process, operating at around room tempera-

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Table 1  
Components in a single ICR18650 cell

Component	Mass (g)	Approximate value		
		£	Euro	US\$
Cobalt	6.5	0.160	0.245	0.248
Electrolyte	5.0	0.151	0.230	0.232
Aluminium	1.6	0.002	0.002	0.002
Copper	2.8	0.003	0.005	0.005
Lithium	0.8	0.004	0.006	0.006
Other	22.6	–	–	–

ture, and recovering more of the cell contents. Such a process has been under development at AEA Technology Batteries, now based at Culham, Oxon, UK. The process involves the following main stages:

#### 4.1. Mechanical shredding

The cells are posted into an inert, dry atmosphere for mechanical shredding. This avoids exposing the cell contents to water vapour, which would hydrolyse the electrolyte. It also reduces the impact of internal short circuits, which can be violent in contact with oxygen.

#### 4.2. Electrolyte extraction

The liquid electrolyte is dispersed in the pores of the electrodes and separator. However, by immersing in a suitable solvent for a few hours, the electrolyte can be extracted. After separation from the residual solids, the solvent can be recovered by evaporation at reduced pressure, leaving pure electrolyte. Several liquids could be used as the extraction solvent. The main requirements are that the boiling point at reduced pressure is below the lithium salt decomposition temperature ( $\sim 80^\circ\text{C}$ ), and that the material is available in an anhydrous state.

#### 4.3. Electrode dissolution

The electrodes are cast using the PVDF binder dispersed in a solvent. This process can be reversed to recover the electrode particles. The cell pieces are immersed in the solvent, which is stirred, heated to around  $50^\circ\text{C}$ . The binder re-dissolves, separating the electrode particles from the residual copper, aluminium, steel and plastic. The latter can be further separated based on their physical properties, e.g. density, magnetism. The electrode particles are filtered from the binder solution, which is then concentrated to recover the bulk of the solvent for reuse.

#### 4.4. Cobalt reduction

The residual electrode particles are lithium cobalt oxide, and one or more types of carbon. Before they can be reused, they need to be separated further. To avoid adding any

chemicals to the system, electrochemical reduction is used. As the cobalt(III) is reduced to cobalt(II), lithium is released from the solid structure. Oxygen is generated at the counter electrode, giving a (simplified) overall equation of:



The carbon particles increase the electronic conductivity during the electrochemical reaction. Any residual lithium is de-intercalated at the same time. Hydrogen evolution is an unwanted side reaction. Therefore, aqueous lithium hydroxide is used as electrolyte, and the current collector is graphite.

### 5. Process development and optimisation

The individual steps of the process were initially investigated separately, on a small bench top scale. This allowed suitable solvents and operating conditions to be determined. It was then decided to integrate the steps into one demonstration unit. This had a number of advantages:

- proper anhydrous operation where required;
- any cross-contamination between process steps revealed;
- consideration of preferred construction materials;
- control and safety issues resolved;
- identification of areas requiring further scale up effort.

Operation of the demonstration unit is continuing, and the latest performance results will be presented at the conference.

### 6. Market size

There are two sources of raw material for recycling; manufacturing waste, and collected used consumer batteries. Fig. 1 shows an estimate of the quantities of both types over the next few years. The numbers are based on predicted sales of ICR18650 cells, with 10% manufacturing waste, an operating life of 3 years, and a 50% collection efficiency.

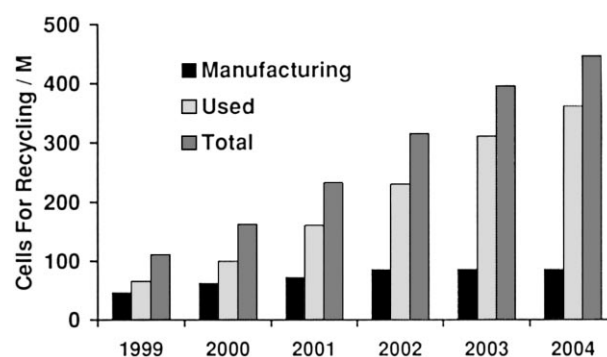


Fig. 1. Estimated numbers of lithium ion cells for recycling.

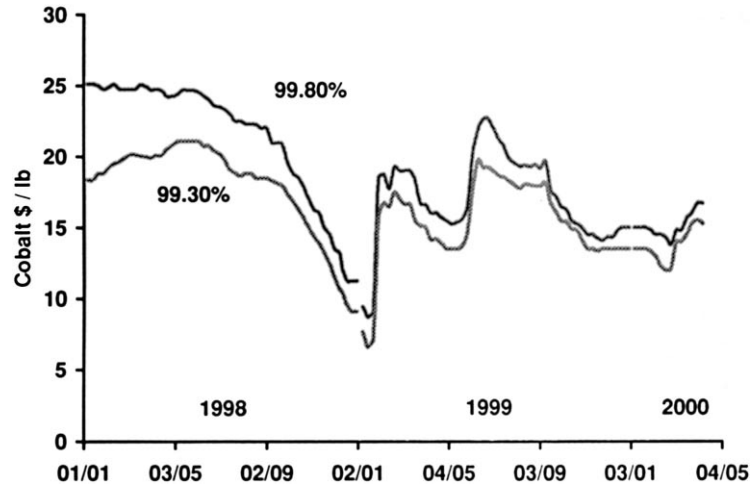


Fig. 2. World cobalt prices through 1998 and into 1999.

## 7. Pack disassembly

Manufacturing waste is comparatively free of contamination, and generally localised. However, the predominant waste source is used batteries. All consumer lithium ion cells are sold as battery packs. Therefore, it is necessary to be able to disassemble the packs, and extract the cells. Unfortunately, there are already several hundred different pack sizes in existence, which makes fully automatic disassembly difficult. However, it is likely that a semi-automated process can be developed. The key safety requirement is to avoid rupturing the cell during any cutting process. Contamination with other battery chemistries can be minimised by taking advantage of the non-standard cell sizes of lithium ion cells.

## 8. Commercial opportunities

Given the projected numbers of cells available for recycling, and the value per cell of the contents, there is a significant market opportunity for recycling lithium ion cells and batteries. Unlike some recycling processes, the

bulk of the income comes from the sale of the products, rather than from charges to the waste disposers. It seems unrealistic to charge the latter more than the costs of disposal through incineration or landfill.

A preliminary design has been produced for an operational plant with an annual throughput of around 10 million cells. This has enabled the capital and operating costs to be estimated. With further assumptions on income, even a single plant shows a solid return on investment. The main complication is the variation in world prices of the recycled products. For example, Fig. 2 shows the recent price of two grades of cobalt. During the operating life of a plant, the prices can be expected to cycle up and down several times. This will obviously effect the degree of profitability, but only the degree, and not the fundamentally profitable nature of the process.

## References

- [1] W.J. McLaughlin, in: Proceedings of the International Seminar on Battery Waste Management; US Patent 5,345,033.
- [2] D. Smith, in: Proceedings of the 10th International Seminar on Battery Waste Management, 1998.