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In the Lab

Alternative Recycling Process for Li-ion Batteries: Molten Salt Approach

Introduction

Dr Ruth Carvajal-Ortiz's current research centered around the innovation in energy storage. She has a special focus on the characterisation of materials, molten salts and their potential applications to several industrial processes, such as metal production and recovery. Currently a research fellow at Coventry University, Dr. Carvajal-Ortiz is in charge of the molten salts recycling work package of the CALIBRE consortium, a circular economy project for automotive lithium-ion batteries.

Ruth's academic and industrial background includes electrochemical characterisation techniques (e.g. voltammetry), corrosion in metals under hydrothermal conditions and synthesis and characterisation of catalysts. She obtained her doctoral degree from the University of Manchester, where she worked designing and testing an *in-situ* molten salt electrochemical oxidation cell to measure hydrogen diffusion in zirconium alloys[1]. Prior to her doctoral studies, Ruth worked with corrosion in metals involved in nuclear applications, during her M.Sc. and as a research chemist at Trent University in Peterborough, Canada. The main project at the Trent University's supercritical water lab was part of the GEN-IV Nuclear Reactors investigations. The aim of the project was to understand the corrosion behaviour of stainless steel in hydrothermal and supercritical conditions.[2-4] The project included colaborations with the Canadian Nuclear Society in Chalk River, Ontario. Ruth's background also includes synthesis and characterisation of catalysts such as titania, used in biofuels.

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About the Research

The demand for lithium-ion batteries (LIBs) has substantially increased during the past years, and is forecasted to grow annually on a global basis by almost 66% by 2025[5] for electric vehicles (EV) alone. This increases the need for an efficient and sustainable recycling process and circular economy system [6]. Coventry University and several battery and automotive companies are working together on a project to provide an achievable and effective way to recycle Li-Ion batteries. CALIBRE (Custom Automotive Lithium Ion Battery Recycling) is a new consortium that covers several stages (Figure 1), from ageing and end-of-life assessment to chemical/molten salt recycling and materials regeneration. Additionally, the project includes a mechanical separation and material recovery process at pilot scale, reuse and life cycle assessment (LCA).

A molten salt recycling process is part of the chemical recycling package. This process provides a novel approach that uses common molten salts as electrolytes and reaction media. The main advantage of the molten salt usage is their performance versatility which, given the multiple choices of battery electrode chemistries that are in the market presently, can be employed as an improvement to current methods.

For the study, different eutectic mixtures of molten salts[7-8] (e.g.: sodium, potassium, lithium and calcium borates and chlorides, sodium and potassium carbonates) are tested to provide an optimised alternative or a shortcut to the hydrometallurgical, pyrometallurgical or even biometallurgical recovery of metals (i.e.: Co, Ni, Mn). This approach takes advange of the salts' electrochemical and solubility properties. Initially, a two-phase molten salt system composed of sodium borate (Na_2O-$2B_2O_3$) and sodium chloride (NaCl) was employed to evaluate the feasibility of metal recovery from mixed feeds of oxides of cobalt, manganese, copper and nickel mixtures and virgin cathode materials (e.g.: NMC 111) by electrodeposition. The process operates within a temperature range of 800-900℃, where both salts are in liquid state. Amietszajew et. al, 2016 reported 98-99% metal purity for single metal oxides deposited using the process described (Figure 2).[9-10]

The system has demonstrated stability and opens up the possibility to be used together with other metal recycling sources and processes. Additional insight into the environmental impact of the pilot scale process such as its carbon footprint and its efficiency are also assessed. The alternative method presented, might solve some of the issues related to the hydrometallurgical methods currently used by recycling the industry, including significant water waste, sulphate by-products and toxic acids use that are detrimental to the environment. Furthermore, the method developed is inclusive of a range of metals, which is of high importance considering the growing and future complexity of the battery waste stream and the need for the world to recycle the materials essential for it, while at the same time reducing greenhouse and pollutants gas emissions.

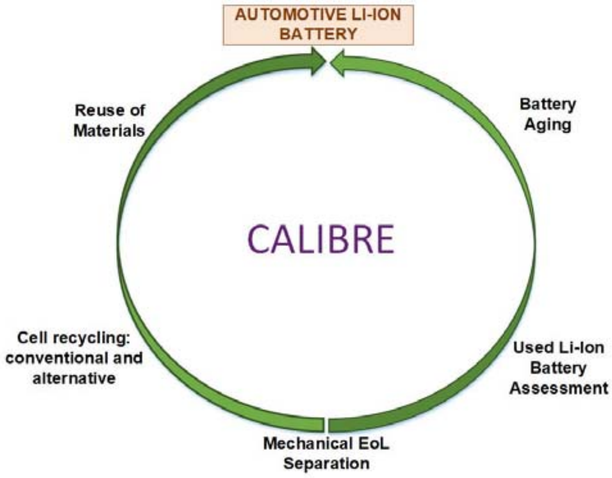

Figure 1. Li-ion battery recycling circular economy project, CALIBRE scheme.

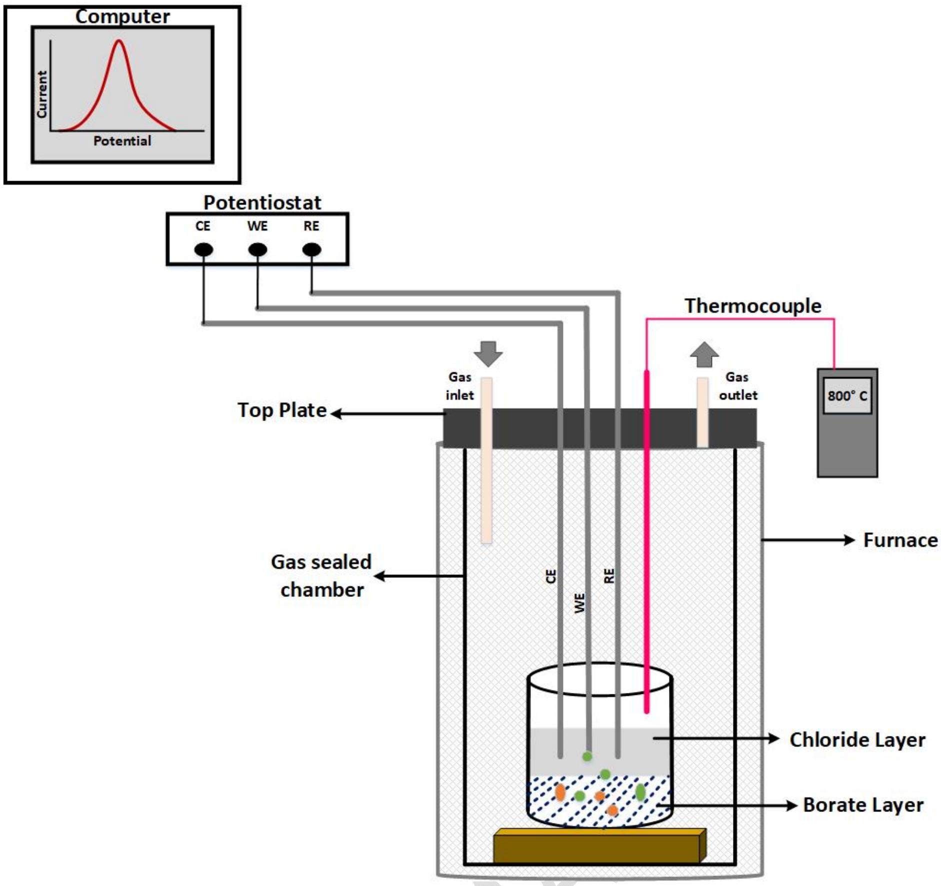

Figure 2. Molten salt electrochemical cell scheme. Cathode and anode reactions during the process of metal deposition.

$$\textbf{Cathode}: Me^{x}_{(l)} + xe^{-} \rightarrow Me_{(s)}$$

$$\textbf{Anode}: Cl^{-}_{(l)} \rightarrow \frac{x}{2} Cl_{2(g)} \uparrow + xe^{-}$$

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