

# Recovery of waste cobalt from lithium-ion batteries and utilization as an electrocatalyst for oxygen reduction and hydrogen evolution reaction

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Hydrogen will play a crucial role to foster the Green Revolution. In particular, electrochemistry to produce and use hydrogen will be paramount. Electrochemical technologies that produce hydrogen are named water electrolyzers (WE). The technologies instead capable of producing electricity through hydrogen oxidation are named fuel cells (FCs). The most performing WEs and FCs are based on a proton exchange membrane (PEM) and the operations are in acid environments. However, in these conditions, the electrocatalysts and other components within the device are based on platinum group metals (PGMs) and critical raw materials (CRMs) skyrocketing the cost and impeding large-scale commercialization. Therefore, solutions need to be found. Importantly, in alkaline environments, PGM and CRM can be substituted with less noble and more abundant materials on the Earth's crust.

In parallel, also batteries will play an important role in the Energy Transition. At the moment, battery technologies are more advanced compared to hydrogen technologies and are more widespread worldwide. Particularly, lithium-ion batteries (LIBs) will have central interest due to the high energy density and the possibility of being recharged several times. However, LIB utilizes materials containing lithium and cobalt that are considered CRMs. Recycling is then a must. Specifically, attention is devoted to the recovery of cobalt.

In this work, we recover the waste cathode of a lithium-ion battery containing cobalt and we used it as electrocatalysts for oxygen reduction reaction (ORR) and hydrogen evolution reaction (HER) in alkaline media. Three materials were investigated and compared: 1) waste cathode "as it is" removed from a spent LIB (Waste LCO); 2) lithium cobalt oxide commercially purchased (LCO), 3) waste LCO subsequently treated with choline chloride: citric acid (1:1) DES with subsequential post thermal treatment (ChCl:Citric) [1]. Initially, the three materials were fully characterized microscopically and spectroscopically. After this initial screening, Waste LCO, LCO and ChCl:Citric were mixed separately with Ketjen Black (KJB) at different percentages of 10% wt and 50% wt.

Electrocatalytic activities of the three materials were studied using a rotating disk electrode (RDE) in nitrogen saturated 1M KOH electrolyte for HER and a rotating ring disk electrode (RRDE) in oxygen saturated 0.1M KOH electrolyte for ORR. For both HER and ORR, 50:50 KJB:Co-based materials were more performing compared to the 10:90 ratio.

For HER, Waste LCO and LCO showed an overpotential of 277 mV at 10 mA cm<sup>-2</sup>. Considering ORR, the most performing electrocatalyst was ChCl:Citric 50-50 with catalyst loading of 0.6 mg cm<sup>-2</sup> showing a promising onset potential ( $E_{\text{onset}}$ ) of 0.85 V (vs RHE) together with a half-wave potential ( $E_{1/2}$ ) of 0.76 V (vs RHE). Waste LCO 50-50 had an  $E_{\text{onset}}$  of 0.83 V (vs RHE) and  $E_{1/2}$  of 0.7 V (vs RHE). [1]

No doubt that the electrocatalytic activities of the derived electrocatalysts are inferior compared to scarce and economically unviable PGM-based electrocatalysts but prospective endeavors could improve the electro-kinetics of the waste-derived electrocatalysts. Such approaches can give rise to new opportunities for green and cost-effective energy within the framework of the circular economy.

[1] S. A. Mirshokraee, M. Muhyuddin, R. Morina, L. Poggini, E. Berretti, M. Bellini, A. Lavacchi, C. Ferrara, C. Santoro. Upcycling of Waste Lithium-Cobalt-Oxide from Spent Batteries into Electrocatalysts for Hydrogen Evolution Reaction and Oxygen Reduction Reaction: A Strategy to Turn the Trash into

