The battery recycling process in Umicore:

The Umicore process for recycling lithium-ion batteries is a prominent method in the sustainable management of battery waste, particularly from electric vehicles (EVs). One of the main advantages of the Umicore process is its high recovery rates for critical metals, which are essential for manufacturing new batteries This process, which integrates heating and chemical techniques, is designed to efficiently recover valuable metals such as cobalt, nickel, and lithium. The process is a combination of a pyro and hydrometallurgical process. After batteries are discharged and manually dissembled, they will be sorted. In this section, the relatively valuable fractions like electronics and cables are separated and the disassembled battery cells are mixed with limestone ($CaCO_3$). Reducing agents like (Al and Zn) are also added. Spent batteries comprise 30-50% of the feed into the smelter. This percentage is necessary in order to produce an alloy with sufficient content of Co and Ni.

This material enters the preheating zone where the temperature increases slowly to 300 and the electrolyte evaporates, so that the risk of explosion will be minimized. In the second zone, the temperature rises to 700, where the plastic components and the binders decompose and partly evaporate. In this exothermic process, the hot gases which are rising from the bottom of the furnace are heated up. As a result, there is a need to address the environmental impact of the emissions generated during the smelting process, which could be reduced by implementing advanced emission control technologies These hot gases rise into the preheating zone, cool down and meet the electrolyte vapors to the flue gas treatment section. When gases reach the top of the furnace, they are at a temperature between 250 and 700, however, all the components are preferred to be kept at gaseous condition at the post-combustion stage. In order to prevent the condensation of the evaporated species, a plasma torch is used [1]. Such a torch provides an increase in gas enthalpy with a limited increase in gas volume. At this stage, halogens can be captured by injecting selected products via the plasma torch or directly into the combustion chamber [2]. Toxic halogens, produced by the decomposition of the electrolyte salt $LiPF_6$ and the binder PVDF and also volatile organic compounds are captured by the injection of CA, NA and ZnO in the post combustion chamber. The temperature of the flue gas leaving the furnace is increased to 1150 by a plasma torch before entering the post-combustion chamber. After the post-combustion chamber, the gases are quickly cooled down to a temperature below 300° C. by injecting water vapor. This avoids the recombination of organic compounds with halogens and the formation of dioxines and furans.

For 1 ton of batteries, 5000 megajoules of heat is needed for the smelter and gas clean-up [3]. Since 1kWh is 3.6 MJ, therefore the energy to supply for the furnace would be 1389 kWh. If we consider a household usage of energy 900 kWh, this amount would be equivalent to the consumption of a household for about 1.5 months. ¨

The third zone is the smelting and the reduction zone. In the smelting phase, the shredded battery material is fed into a high-temperature furnace. The intense heat causes the formation of a metal alloy that contains cobalt, nickel, and copper, while other elements like lithium, aluminum, and manganese form a separate slag. There is a regulated flow of pre-heated air at 500, which is injected into the bottom of the shaft furnace. Here, part of the alloy goes to the slag which is in oxidized state and consists of Li, Al, Si, Mn, Ca and residual Fe. The second part is the oxidized state which contains Cu, Co, Ni and Fe. Depending on the battery casing which can be Al (which acts as a reducing agent) or steel (which does not act as a reducing agent) the amount of carbon used for reducing the cathode metals differs. The smelting process takes about 455 minutes (7-8 hours) and results in the following metal recovery rates 92.8% for Cu, 99.0% for Ni, 64.5% for Fe, and 94.0% for Co. The fraction of iron recovered in the alloy, is the least in comparison with its amount in the feed. This is due to the fact that Fe together with other slag formers like Li, Al and Mn, end up in the slag phase, in the form of metal oxides.

The slag from Li-ion batteries could be recovered through hydrometallurgical approaches, but it is not currently economical. As a result, the slag is sold as an additive for construction material. After the recovery of Fe, Cu, Ni and Co to the greatest extent possible and delivering it in an alloy form, the rest of the elements end up in the hydrometallurgical section.

After the formation of a preliminary alloy of Co, Ni, Fe and Cu, these elements have to be separated and brought back to the production site. This separation takes place through a hydrometallurgical process. The alloy is crushed and leached with H_2SO_4 solution to form metal sulfate salts which are dissolved in the aqueous solution.

Cu = to remove the Cu from the solution, SO_2 is added at elevated temperatures, which leads to the precipitation of CuS nd Cu_2O.

Fe = by using NaOH solution, Fe precipitates as hydroxide.

So far, the main remnants of the preliminary alloy would be Co and Ni ions in solution. This resulting solution is then pumped into a mixer settler, where helps Ni to remain in the aqueous phase and sends Co to the organic phase.

Co and Ni = by using organophosphinic acid, Co goes to the organic phase and Ni remains in the aqueous phase

This separation is done at room temperature and the volumetric ratio between organic solvent and aqueous solution is in the range of 1-5. The pH is also set to around 5.

The selectivity of organophosphinic acid for metal ions is as follows:

$Fe^{3+} > Zn^{2+} > Cu^{2+} > Mn^{2+} > Co^{2+} > Mg^{2+} > Ca^{2+} > Ni^{2+} > Li^{+}$

As a result, Fe, Cu and Mn which have remained as impurities, end up in the organic Co-rich phase, while the aqueous solution which is rich in Ni, will contain Co as an impurity.

The Umicore process represents a robust and sustainable approach to recycling spent lithium-ion batteries. By efficiently recovering valuable metals and minimizing environmental impact, it significantly contributes to the circular economy in the battery industry. Future research and development should focus on optimizing the process, reducing energy consumption, and adapting to evolving market and regulatory landscapes.

1. Latini, D., et al., *A comprehensive review and classification of unit operations with assessment of outputs quality in lithium-ion battery recycling.* Journal of Power Sources, 2022. **546**: p. 231979.
2. Cheret, D. and S. Santen, *Battery recycling*. 2007, Google Patents.
3. Sonoc, A., J. Jeswiet, and V.K. Soo, *Opportunities to improve recycling of automotive lithium ion batteries.* Procedia cIRP, 2015. **29**: p. 752-757.