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Optimized strategies for the recovery of critical raw materials from end-of-life SOFCs

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Scientific context

SOFCs (Solid Oxide Fuel Cells) represent a **highly efficient, fuel-flexible and environmentally friendly technology**, alternative to fossil fuels, that offers a long-term reliable energy supply in the transition towards decarbonization. However, along with some major unmet challenges, especially in terms of life-cycle costs, the full deployment of SOFCs is still hindered by the **lack of efficient, scalable and cost-effective end-of-life (EoL) strategies** enabling the management and valorization of waste products derived from stacks operation and avoiding their landfill disposal.

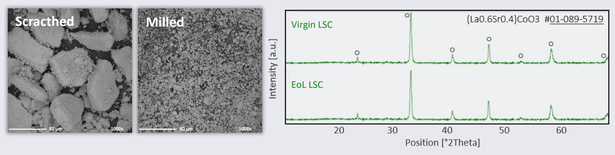
Within this framework, the **BEST4Hy project**, supported by the EU's Horizon 2020 Research and Innovation Programme, aims at developing and validating **new recycling strategies for the recovery of critical raw materials** (Ni, YSZ, La and Co) from EoL SOFCs for re-use in cell manufacturing. At the end of the processes, materials will be validated in terms of **quality and performance** in remanufactured SOFCs and stacks, demonstrating the overall efficiency of recycling.

Objectives of the study

This study seeks to specifically adapt and optimize recycling processes for the **recovery of Ni and YSZ from EoL anode-supported SOFCs**. The recovered materials are required to target fixed acceptance criteria for cell remanufacturing, with special focus on particle size distribution, specific surface area and chemical purity.

Cathode detachment

The detachment of LSC (Lanthanum Strontium Cobaltite) cathode from EoL cells has been carried out through manual **mechanical scratching**.



The recovered LSC powders are repeatedly milled and sieved below 20µm to target the specific acceptance criteria for direct re-using (**14% wt loss**).

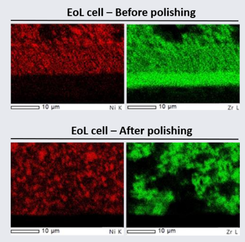
In parallel, the development of specifically adapted processes is under study [1,2,3,4] to selectively recover La and Co in the form of precursors for both closed-loop and open-loop recycling.

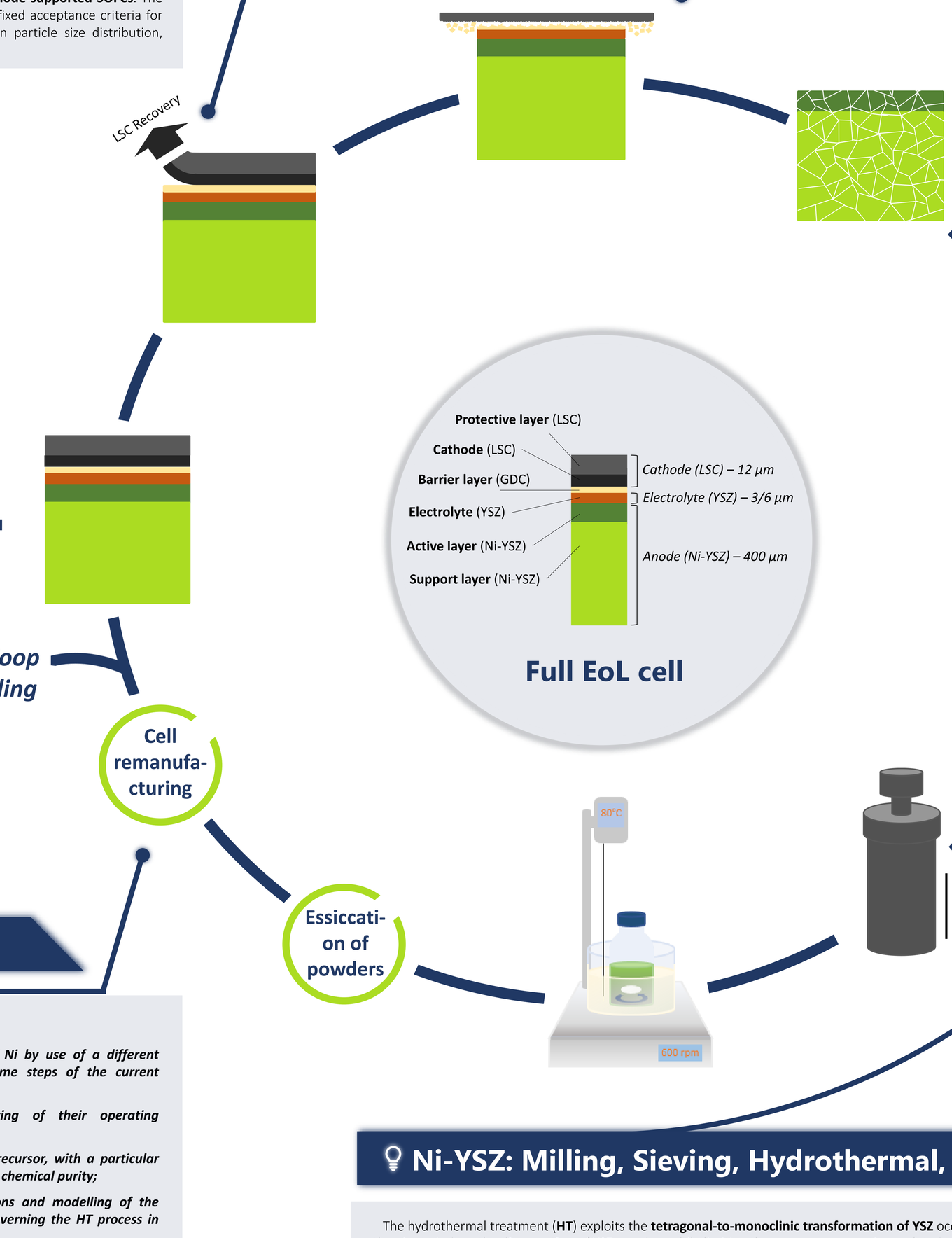
Polishing of residues

LSC residues and GDC barrier layer are successfully removed, leading to a **high purity** of the recovered anode materials. According to EDS mapping, the thin 8YSZ electrolyte is concurrently polished away. The recovery process thus regards the main cell components, i.e. **Ni-3YSZ**.

EDS Elemental Analysis

Before polishing		After polishing	
Element Symbol	Atomic Conc. (%)	Element Symbol	Atomic Conc. (%)
O	58.77	O	27.06
Ce	10.47	Ce	ND
Gd	3.69	Gd	ND
Sr	3.12	Sr	ND
Zr	14.30	Zr	17.36
Ni	6.90	Ni	52.78
Y	2.16	Y	1.51

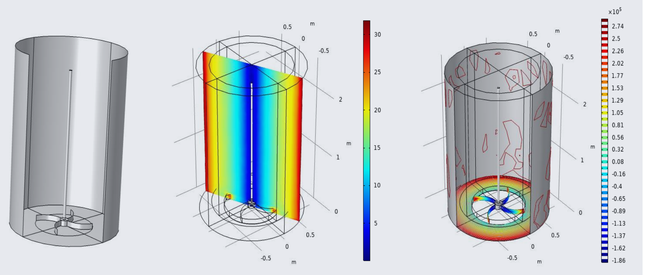




Future work

Perspectives

- *Improving the extraction efficiency of Ni by use of a different extracting agent or by modifying some steps of the current process;*
- *Remanufacturing of cells and testing of their operating performances by our industrial partner;*
- *Reprecipitation of Ni in the form of precursor, with a particular focus on its particle size distribution and chemical purity;*
- *Assessment of the energy consumptions and modelling of the fluido-dynamic and thermal aspects governing the HT process in view of its scaling up to TRL5.*



Ni-YSZ: Milling, Sieving, Hydrothermal, Selective Leaching of Ni

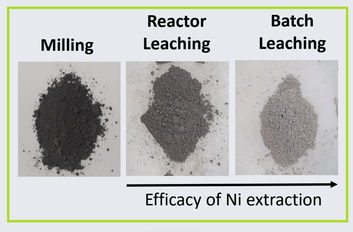
The hydrothermal treatment (**HT**) exploits the **tetragonal-to-monoclinic transformation of YSZ** occuring at high pressures in the temperature range between 200 and 240°C [5], with the aim to induce the **disgregation** of YSZ powders while limiting the energy consumptions related to the milling step.

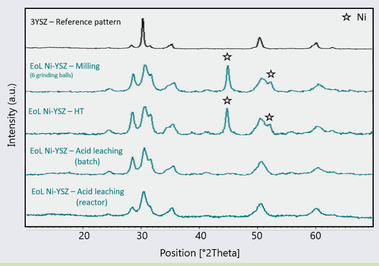
For the **selective extraction of Ni**, the operating parameters (HNO_3 2.2M, 600 rpm, 80°C, 2h) of an **existing procedure** reported for the acid-assisted treatment of Ni-YSZ Cermet [6] have been **adapted and optimized** to extract Ni from the Ni-YSZ anode components of SOFCs, **both in batch and reactor conditions**.

Key Outcomes

Leaching: • Batch conditions – stirring

• Reactor conditions – no stirring





	Milling	HT	Batch Leaching	Reactor Leaching
Specific Surface Area ($m^2 g^{-1}$)	8	13	16	21
Particle size distribution (nm)	Particles in the range of 400-1000 µm (as detected by Dynamic Light Scattering - DSL)			
Ni content (Atomic conc.)	27	20	0.6	1.0
Weight loss after leaching (wt%)	-	-	30	43

ICP Elemental Analysis (ppm) – on the supernatants

	Ni	Y	Zr
HT	1	0	0
Batch Leaching	$8x10^3$	7	180
Reactor Leaching	$3x10^3$	37	10^3

Discussion

- Stirring is fundamental to increase the mass transfer during leaching and thus, the overal efficacy of Ni extraction
- In reactor conditions, YSZ is significantly leached away along with Ni and lost
- Ni residues, not detectable through XRD analysis, have been observed according to EDS evaluations after the leaching process

Reference acceptance criteria

- Specific surface area – 12-20 $m^2 g^{-1}$ ✓
- Particle size distribution of YSZ – 100-600 nm ✓
- Chemical purity of YSZ – above 99% (under optimization)

References

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