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Recycling possibilities for solid oxide fuel and electrolyzer cells

Motivation

Alongside renewable power generation and battery storage, hydrogen technologies are essential, enabling a deep decarbonization of the energy system. For the industrial-scale implementation, the demand for high-performance materials will rise drastically. According to the International Energy Agency (IEA) study from 2021, the global installed electrolyzer capacity in 2050 must exceed terra watt level to achieve the paris 1.5 °C climate goal. Beside other viable hydrogen electrolyzer systems (e.g. proton exchange membran electrolyzers; PEMEL), solid oxide cells (SOCs) offer a great application flexibility with energy efficiencies up to over 90 % and will thus become part of the targeted hydrogen infrastructure. Depending on the SOC design, a number of valuable raw materials are required for the fabrication, often including critical elements such as cobalt (Co), strontium (Sr), nickel (Ni) and rare earth elements (REEs). To preserve these materials, recycling strategies are to be developed even at an early market entry stage, with the aim of integrating SOC systems into circular economy.

Strategies

SOC recycling summed up in 4 steps:

i.) Automated, mechanical stack dissection.
ii.) Mechanical separation of fundamental components (metals, ceramics, miscellaneous).
iii.) Further separation of raw material fractions or reconditioning/refurbishing.
iv.) Reprocessing to SOC starting materials or utilization as raw material for other devices.

The challenges in SOC ceramic recycling:

- Recycling of cell material depends on the cell design: most established cell types differ in material quality and quantity.
- Separation of functional layers require more effort since the layers are strongly attached (sintered) to each other.
- High material purity often nescessary (electronical leakage, mixed phase formation can lower the cell performance).
- Harmful/toxic components: nickel oxides, chromium oxides.

Material quantities depending on cell type:

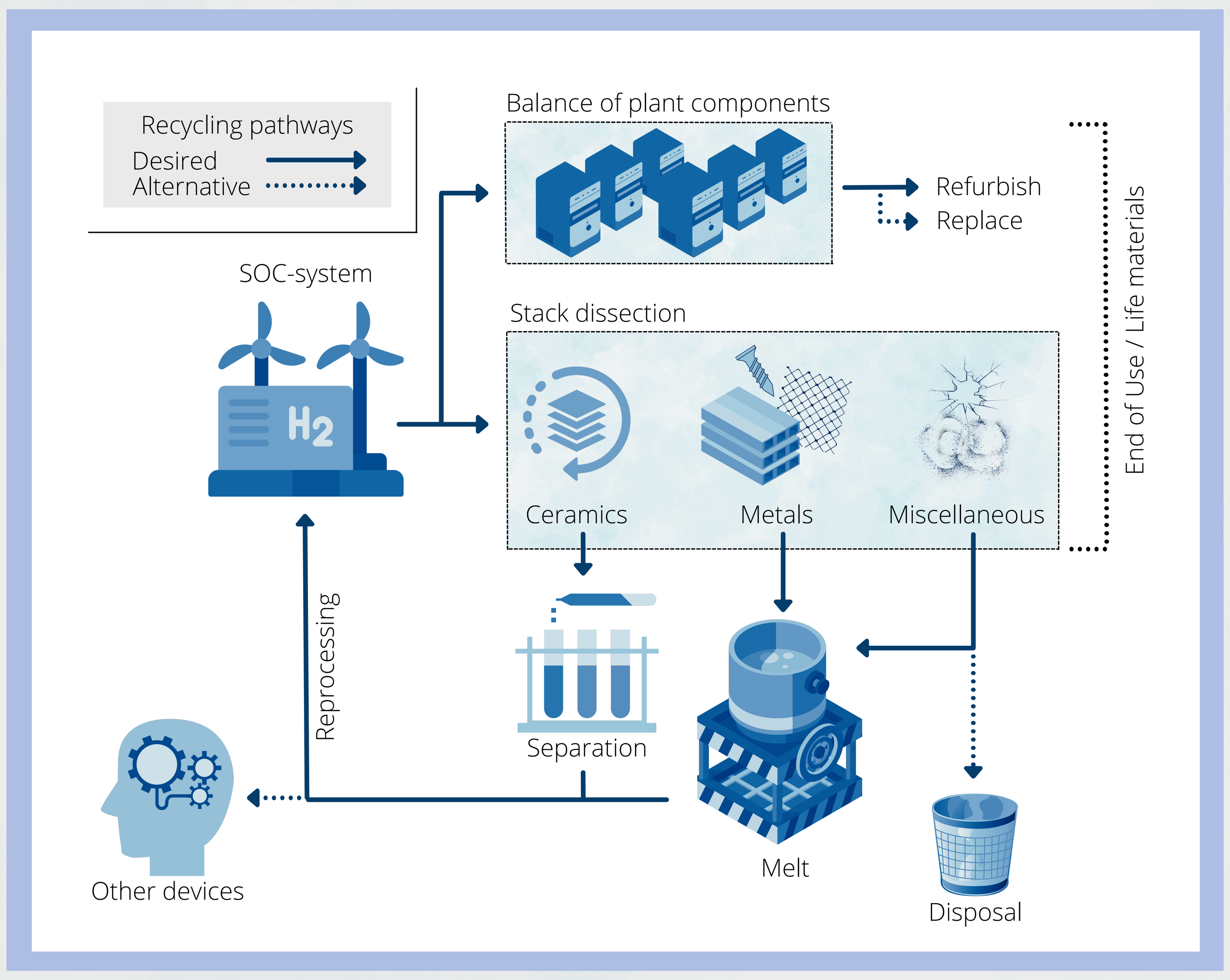

Fig 1. Overview on the desired SOC life cycle and suggested End of use/End of life material pathways.

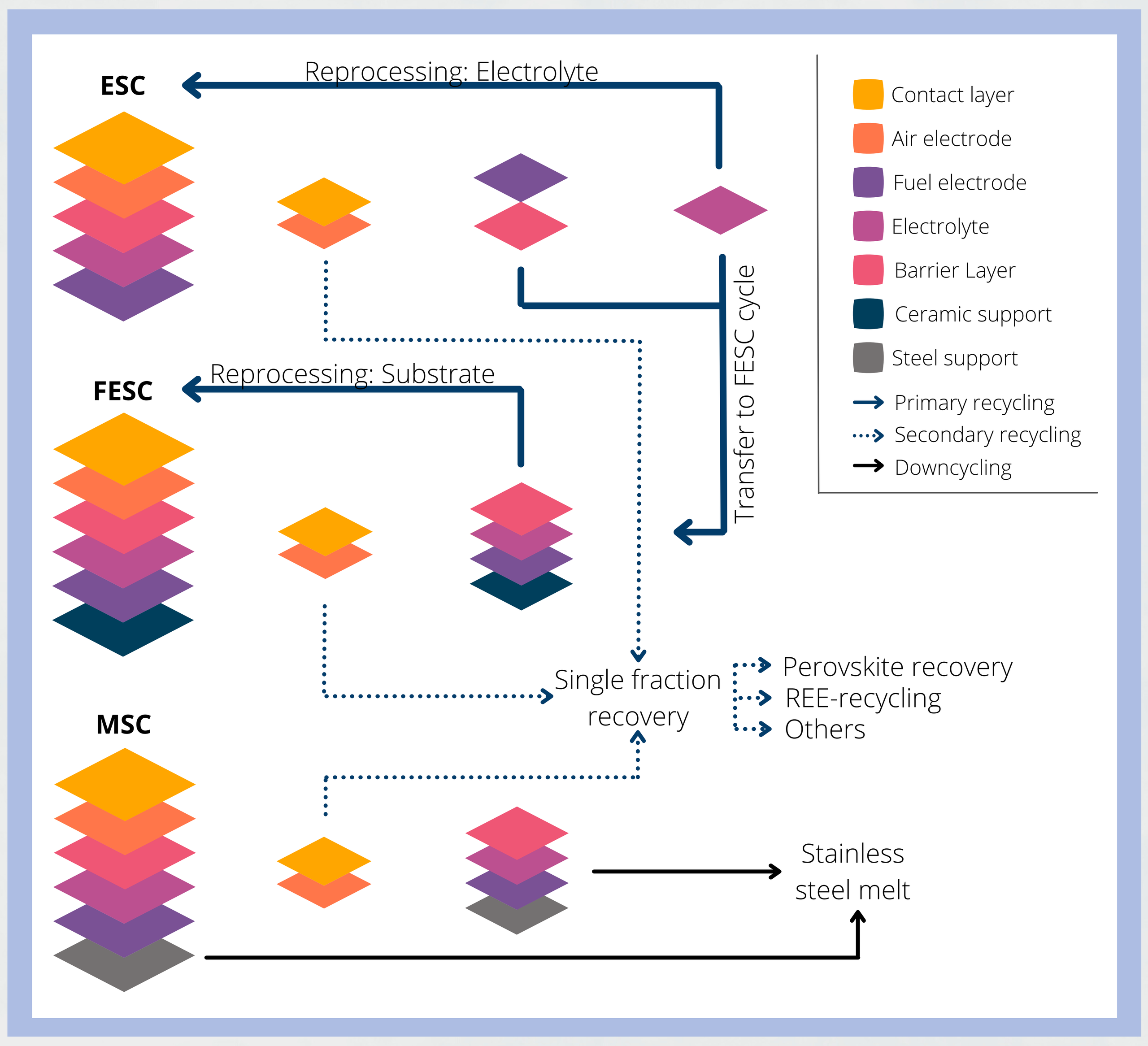

Fig 2. Potential material fluxes for the recycling of different cell types.
ESC electrolyte-supported cells; FESC fuel electrode-supported cells; MSC metal-supported cells

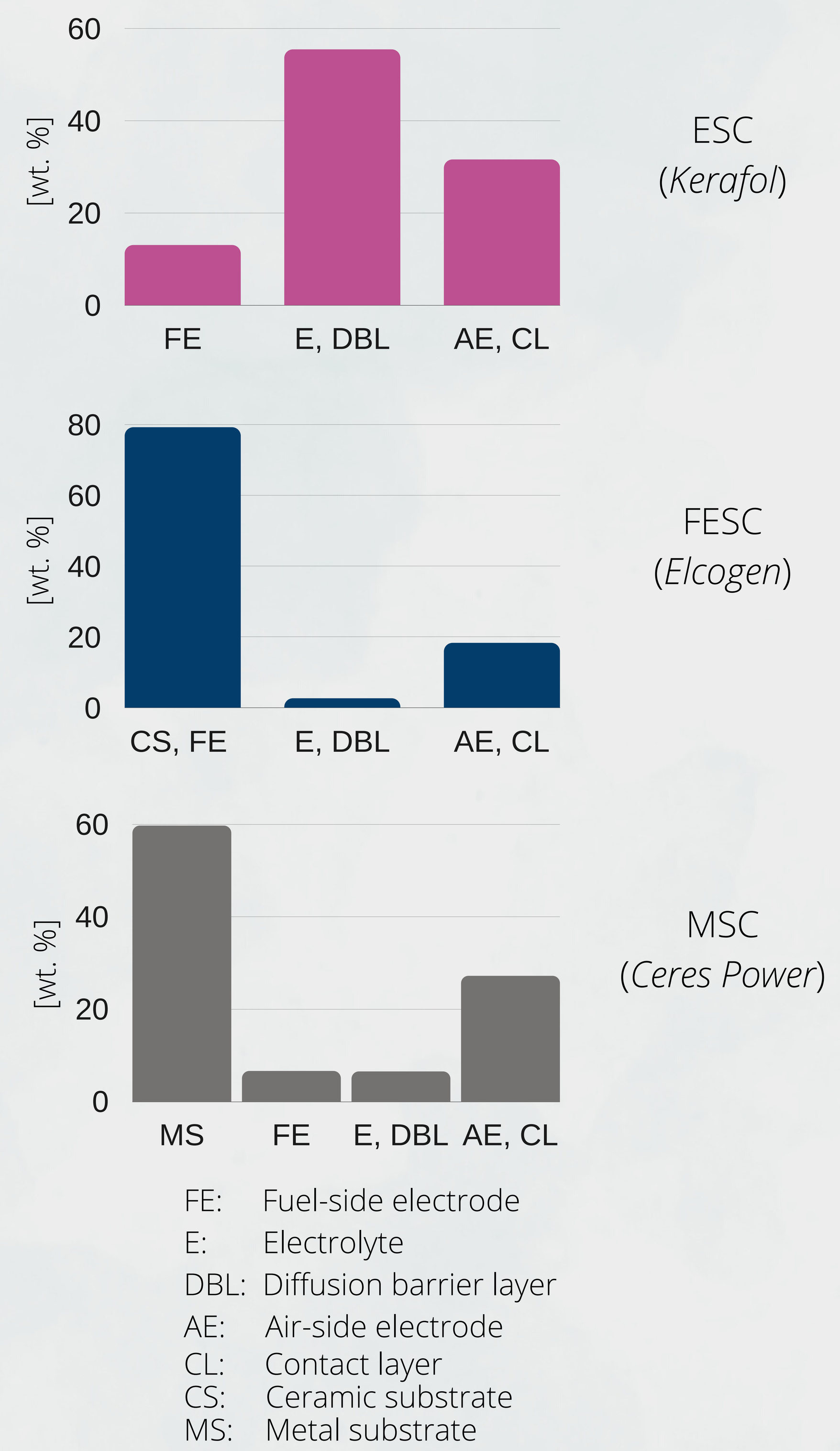

Fig. 3 Assumption of cell weight fractions of three industrial-leading manufacturers.

Experimental

Separation of ceramic functional layers and reprocessing to new slurry/substrate:
The succesful separation of air-side materials from the residual fractions is relevant for each type of cell design and will be the first step in cell recycling. A hydrometallurgical approach can avoid the formation of harmful dust while ensuring a fast and efficient removal of the air-side materials. Alongside suitable solvents, commercial acids such as hydrochloric acid and nitric acid can fulfil the requirements for a selective separation. The hydrometallurgical investigations were performed on fuel electrode-supported cell (FESC) materials from Forschungszentrum Jülich and will thus refer to the FESC cycle in Fig.2. Subsequently, investigations on the reprocessing of ceramic substrate have been performed, starting up with the recycling of old green tape remains to new slurry (Fig. 6).

Methodology development >>>>>>>>>> Feasibility study >>>>>>>>>> Implementation lab-scale >>>>>>>>>> Evaluation >>>>>>>>>> (Upscale)

Optimization of dissolution parameters:

- Statistical design of experiments (Box-Behnken Design).
- Dissolution dependent on T [°C], c [%] and t [min].
- Air-side (lanthanum-strontium-cobalt-iron-perovskite; LSCF) must be dissolved while remaining fractions (> 70 wt.%) are reused for substrate manufacturing.

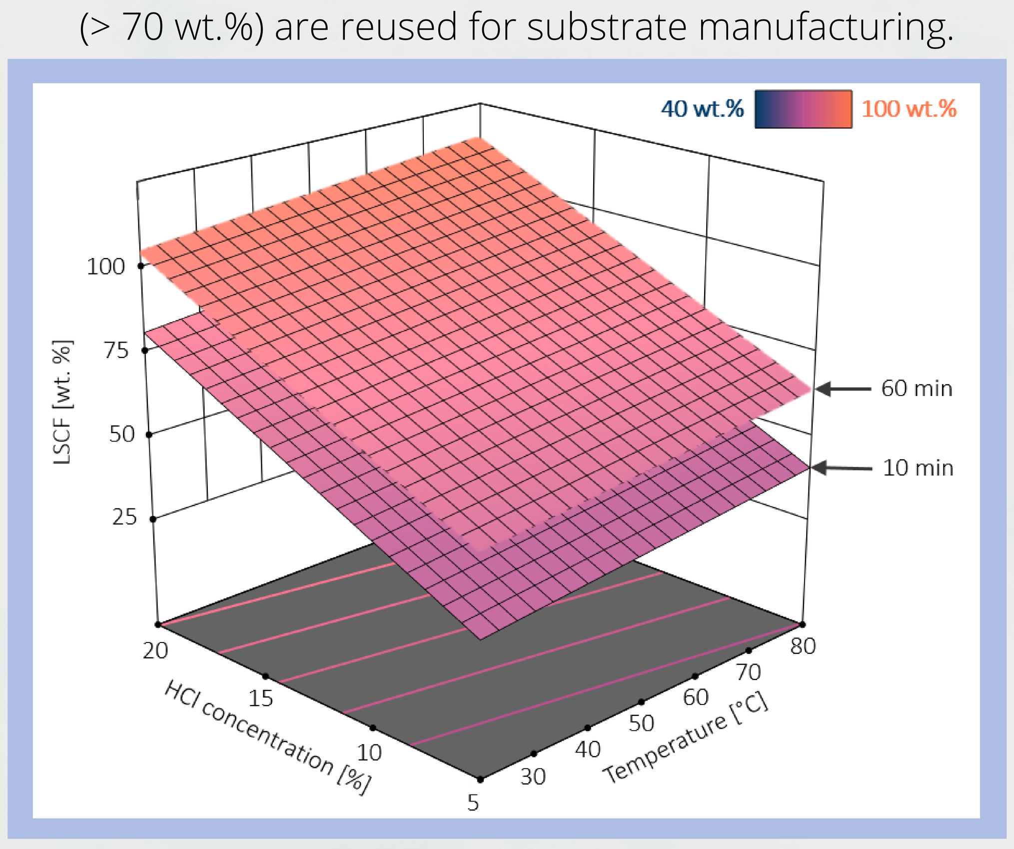

Fig 4. Model of LSCF raw powder dissolution behaviour in hydrochloric acid (HCl).

Benefits of hydrometallurgical separation:

- Accurate separation of perovskite layer possible.
- Main fractions (YSZ, Ni, GDC) can be preserved (prior heat treatment for Ni preservation suggested).
- No development of toxic Ni/Cr dust
- Adaptation to varying layer thicknesses and materials possible.

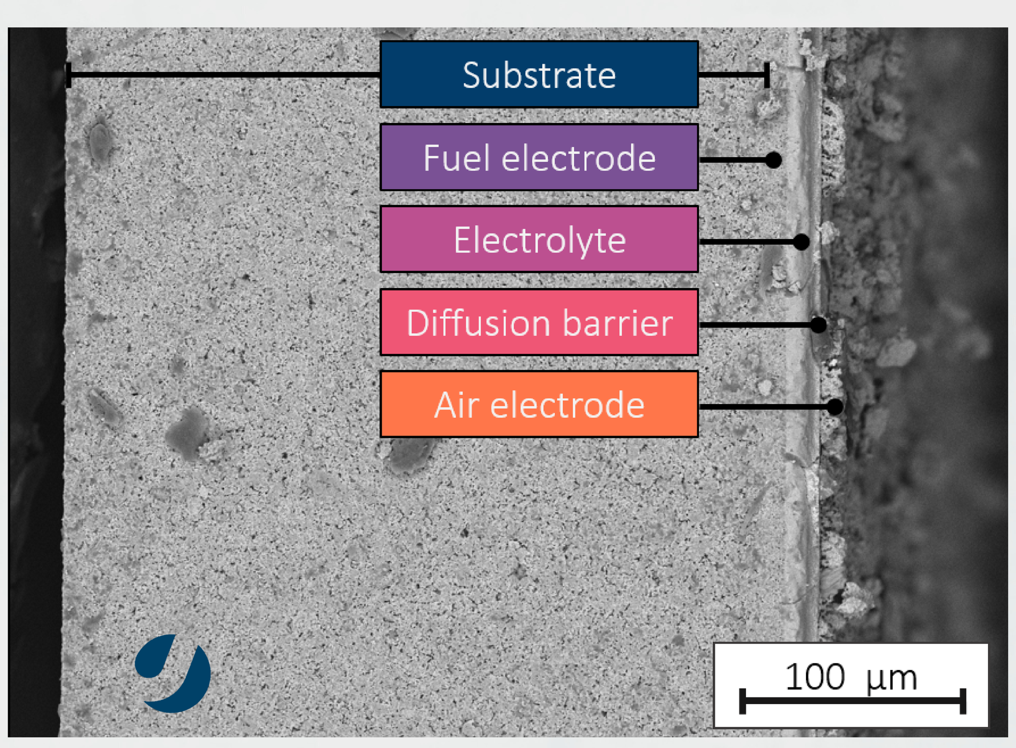

Fig 5. SEM COMPO-mode image of selective etched old cell material (cell crosssection).

Reprocessing of green tape remains to new slurry:

- Slurry parameters (particle size distribution, viscosity, stoichiometry) affect the quality of the substrate properties (durability, electrical and ionic conductivity, liquid and gas transport).
- The preparation of new slurry is matched to the properties of the original slurry so that the substrate properties are maintained.

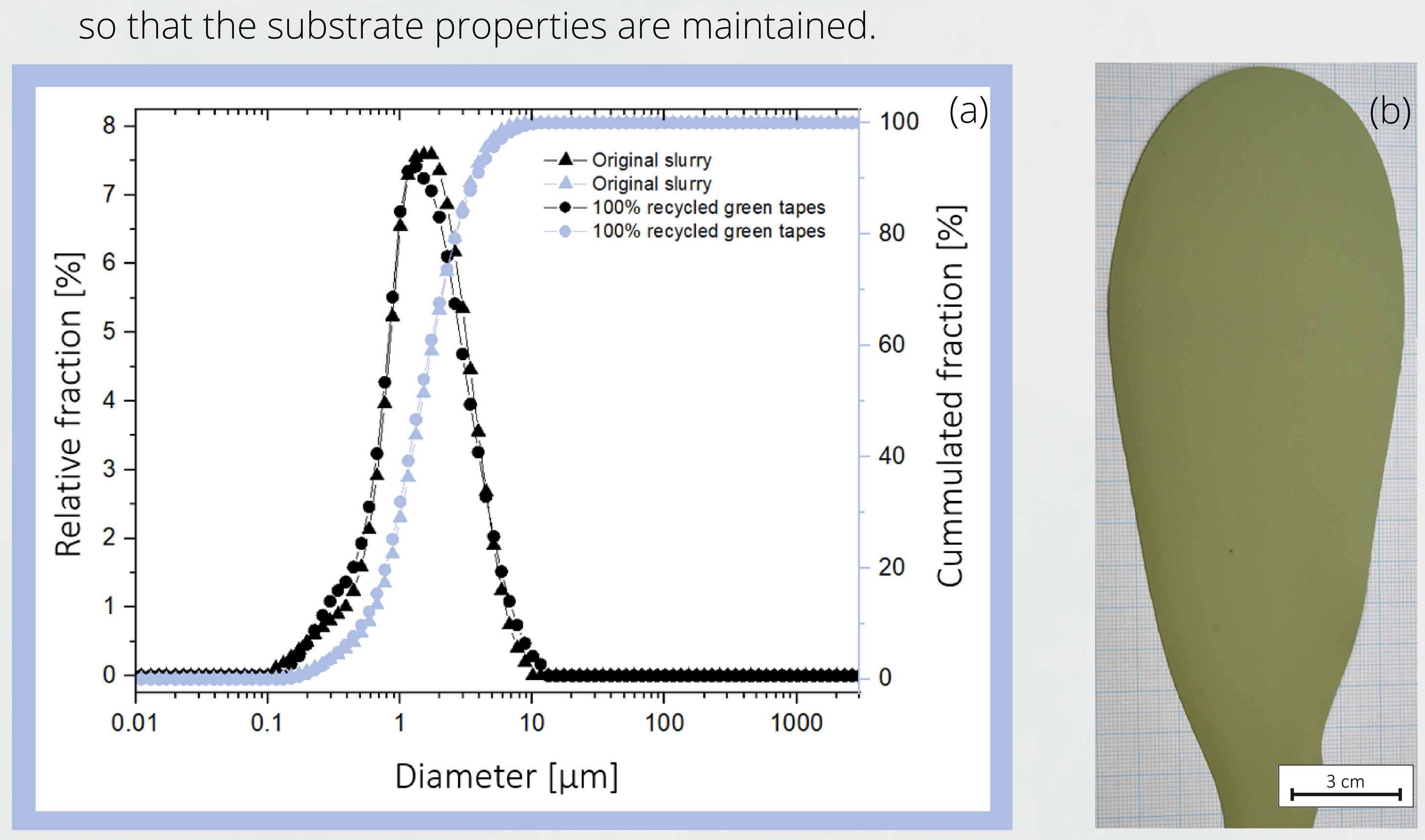

Fig 6. Particle size distribution of the reprocessed slurry (a) and the corresponding casted tape after 1h drying (b).

Outlook

- First experimental results indicate technical feasability of functional layer separation in FESC recycling. The implementation on a larger scale seems to be possible.
- The technical feasibility of returning recovered material fractions to the SOC system without compromising stack performance is still unknown and needs to be investigated.
- Upcoming steps:
  (i.) Processing of sintered cell residues into recycled slurry/ceramic carrier substrate, respectively.
  (ii.) Utilization of dissolved (perovskite-based) fraction - application in SOC or for other devices.
  (iii.) Investigations on other cell types (ESC, MSC design).
  (iv.) Evaluation of economic and ecologic potential (e.g. life cycle assessment studies).

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