CLOSED LOOP RECYCLING OF SOLID OXIDE CELLS

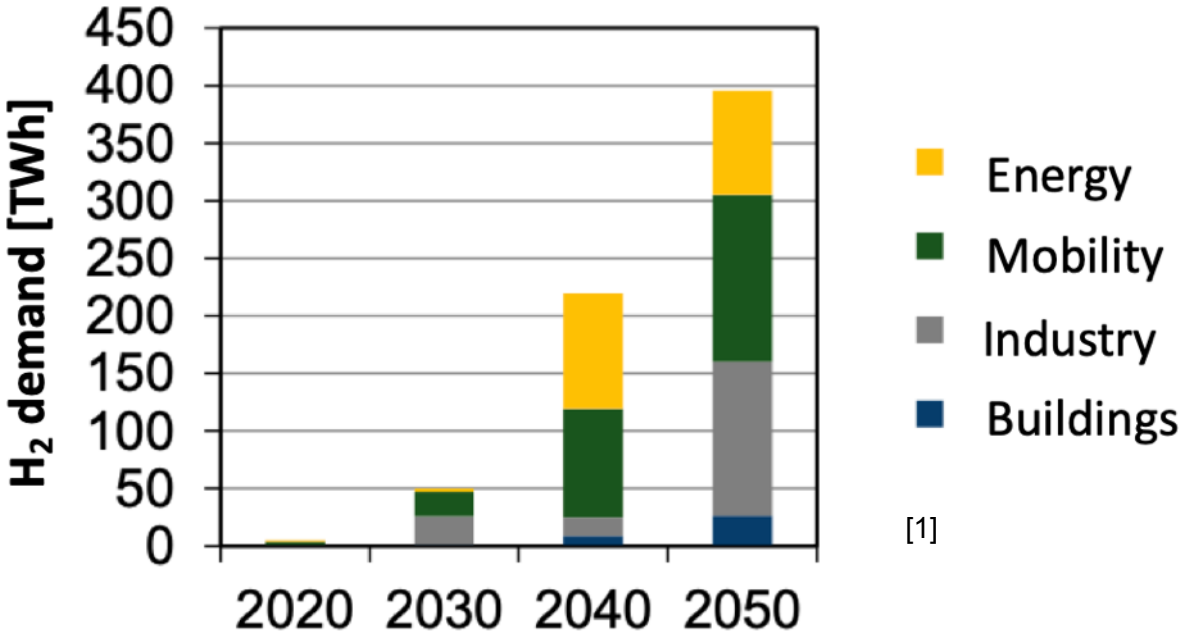
Hans-Walter-Hennicke Presentation Contest 2023

28/3/2023 I MARTIN HILGER I IEK-1, FORSCHUNGSZENTRUM JÜLICH

KERAMIK CERAMICS 2023

JÜLICH Forschungszentrum

SOLID OXIDE CELLS FOR THE “ENERGIEWENDE“



[1]

Growing demand on *green* H_2

[1] Robinius et al., Schriften des Forschungszentrums Jülich (Energy & Environment), 2020

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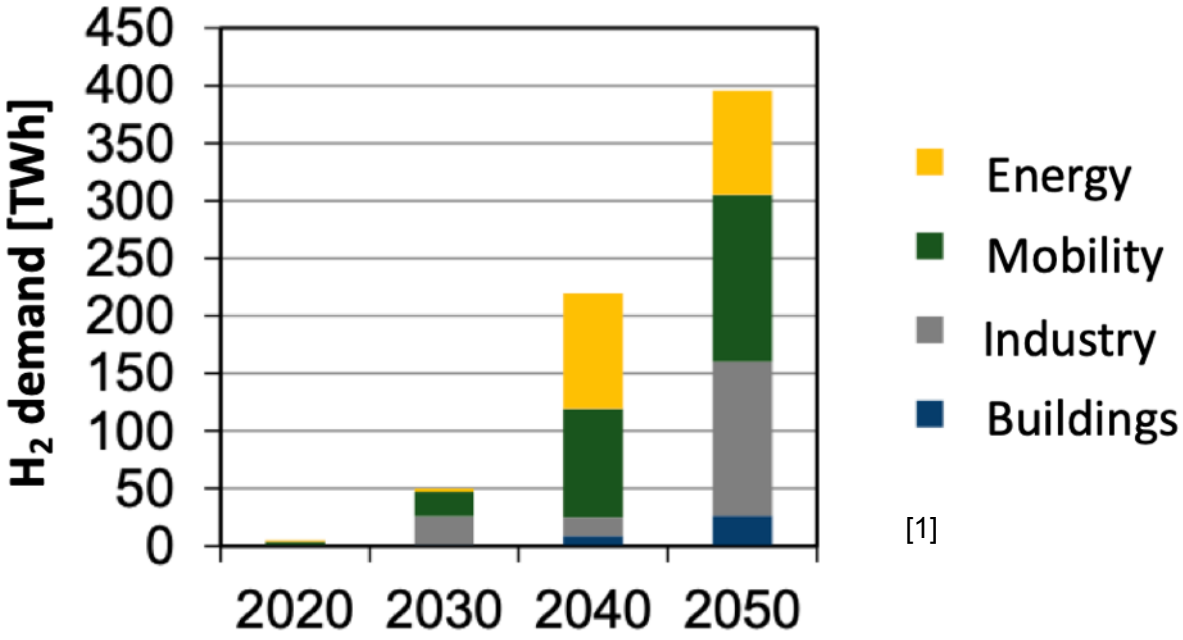

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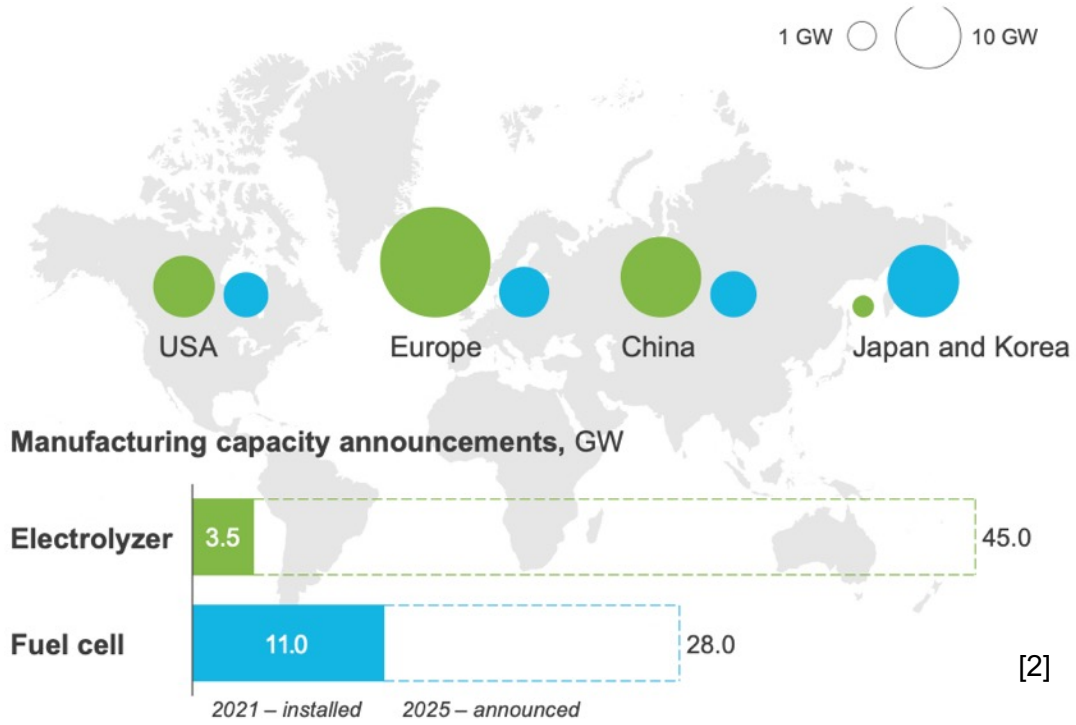

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Growing demand on *green* H_2 → Electrolysis capacities → Resource consumption

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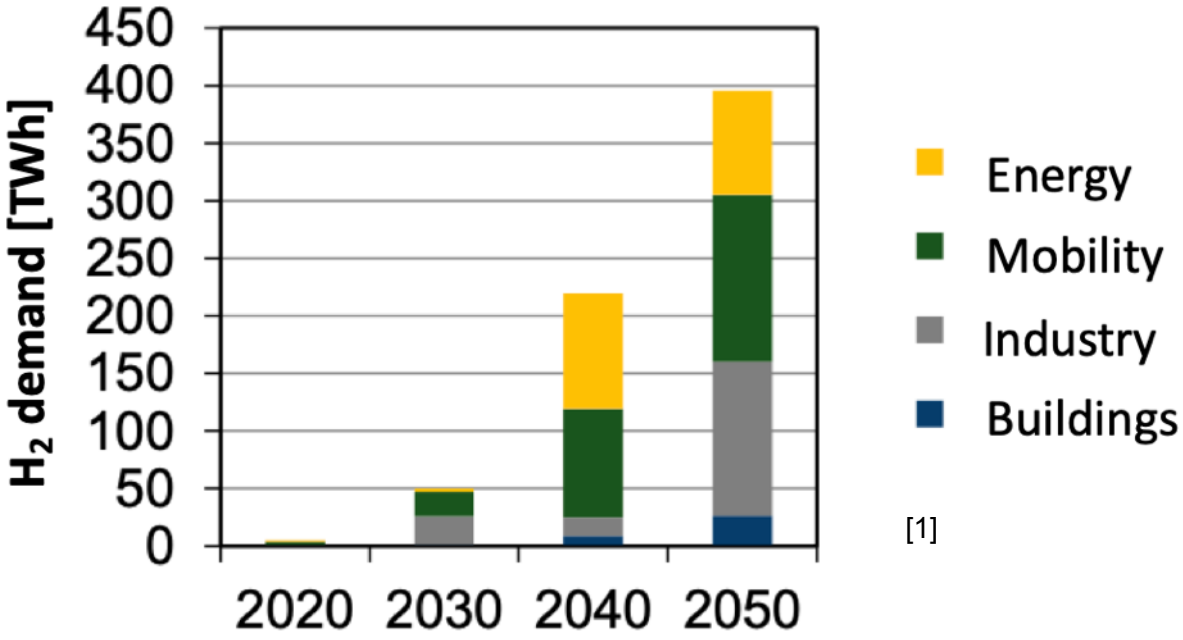

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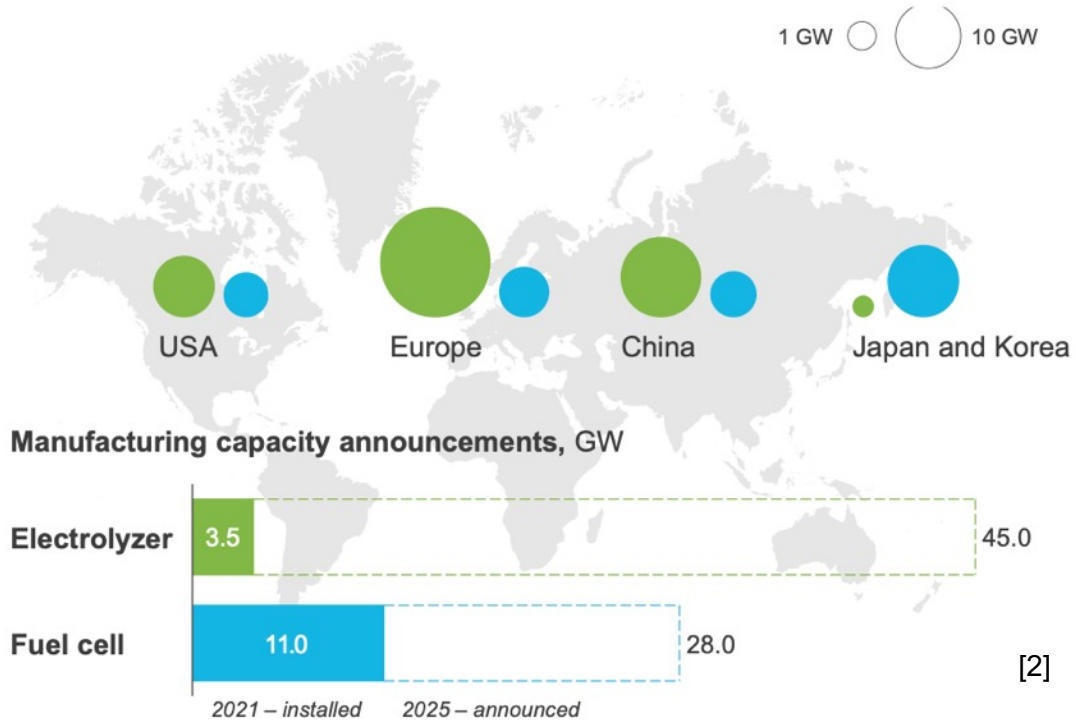

[2]

Growing demand on *green* H_2 → Electrolysis capacities → Resource consumption → **SOCs** → Power-heat-coupling High efficiencies

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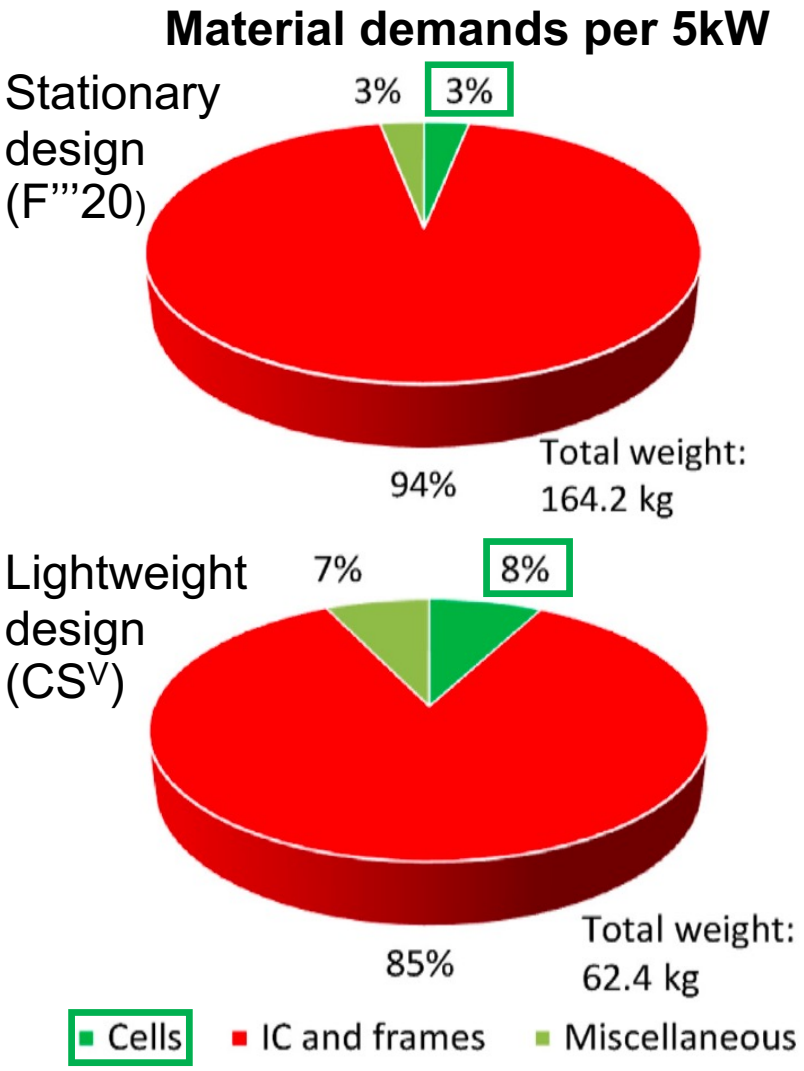
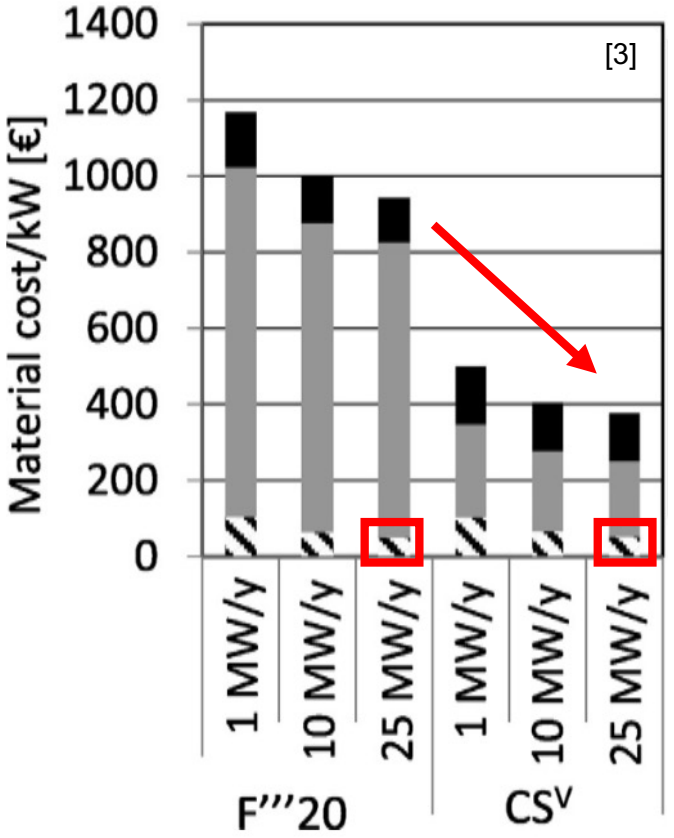
SOCs = Solid oxide cells

WHY CLOSED LOOP RECYCLING OF SOCS?





Lighter stack design

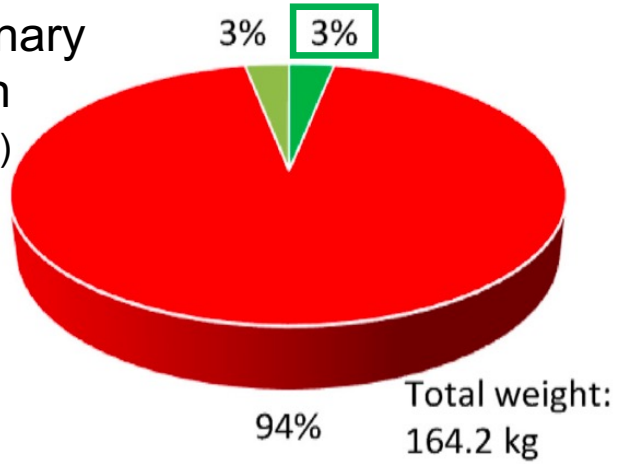
Increasing share of cell in stack weight and costs

[3] Harboe et al., Int. J. Hydrog. Energy, Vol. 45, 2020

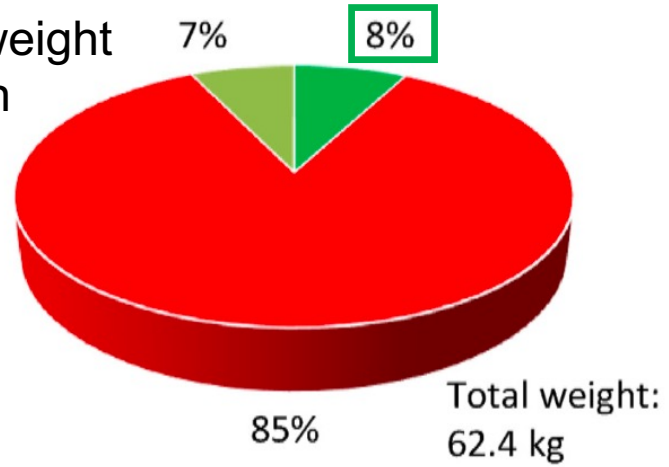
WHY CLOSED LOOP RECYCLING OF SOCS?

Material demands per 5kW

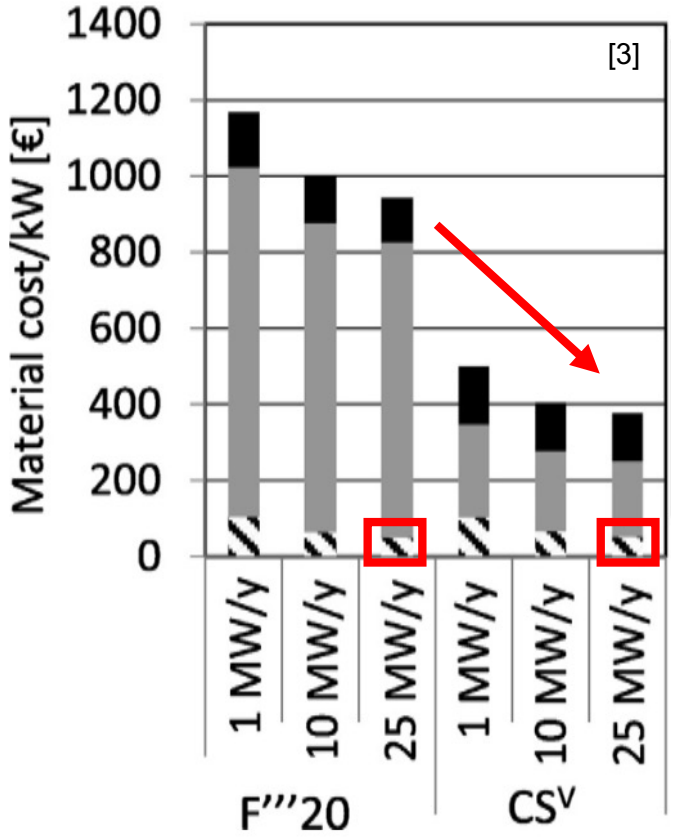
Stationary design (F’’’20)



Lightweight design (CS^V)



Cells ■ IC and frames ■ Miscellaneous



Lighter stack design → Increasing share of cell in stack weight and costs

Limited/ critical resources → REE & PGM Nickel Dependencies

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REE = Rare earth elements PGM = Platinum group metals

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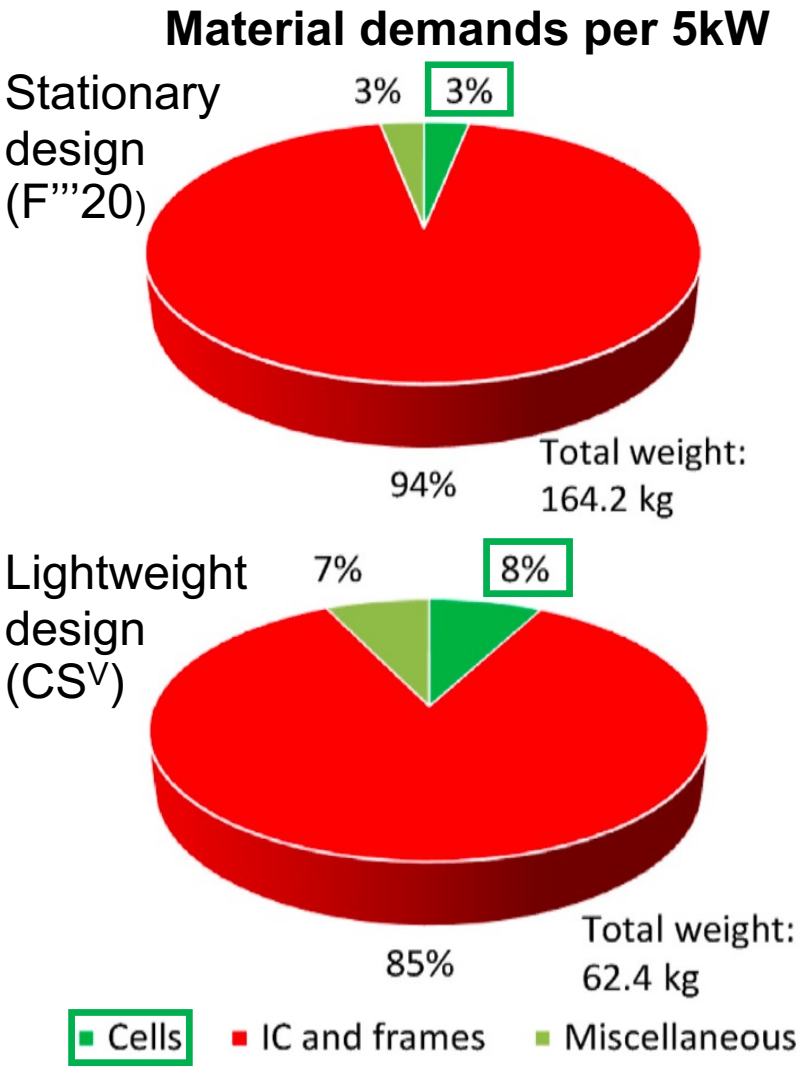

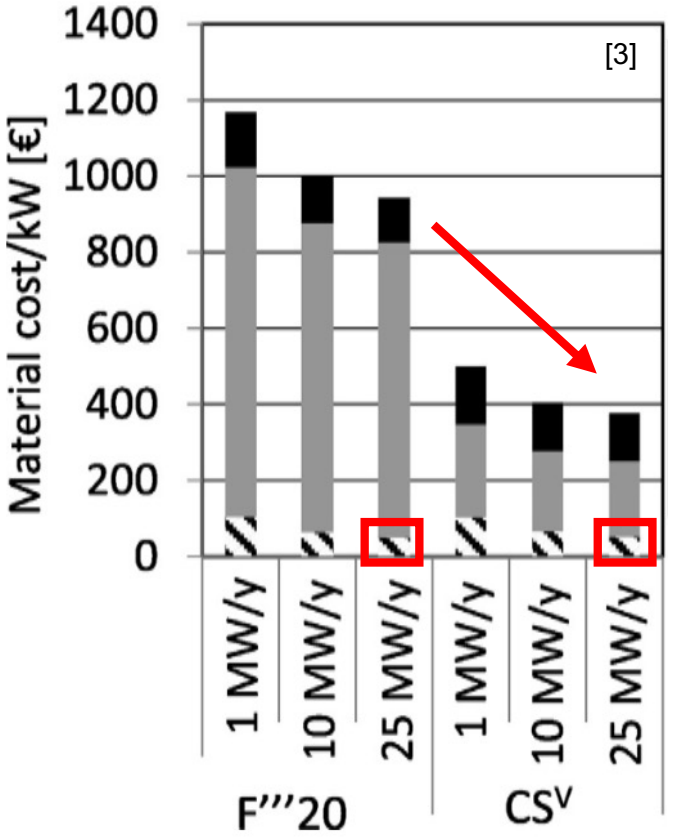

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Rising importance of **closed loop recycling** of cell material

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STRUCTURE OF SOLID OXIDE CELLS (FZ JÜLICH)

Elcogen, Website, 2023

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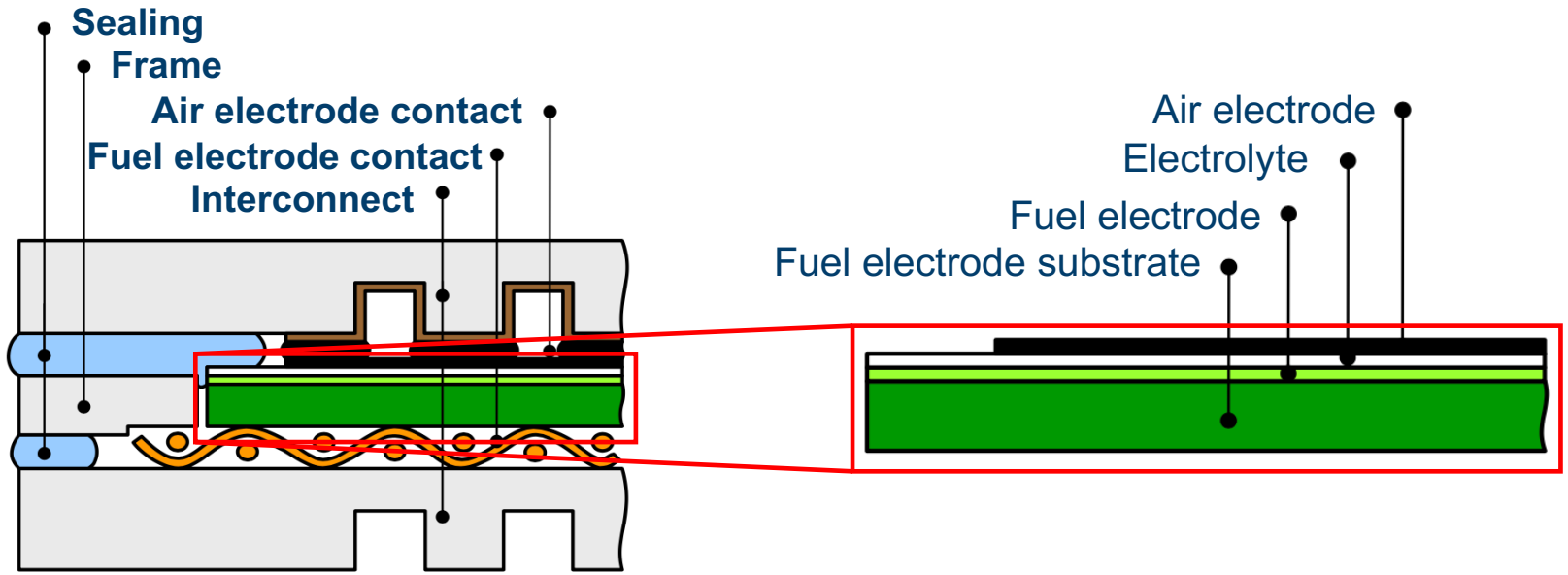

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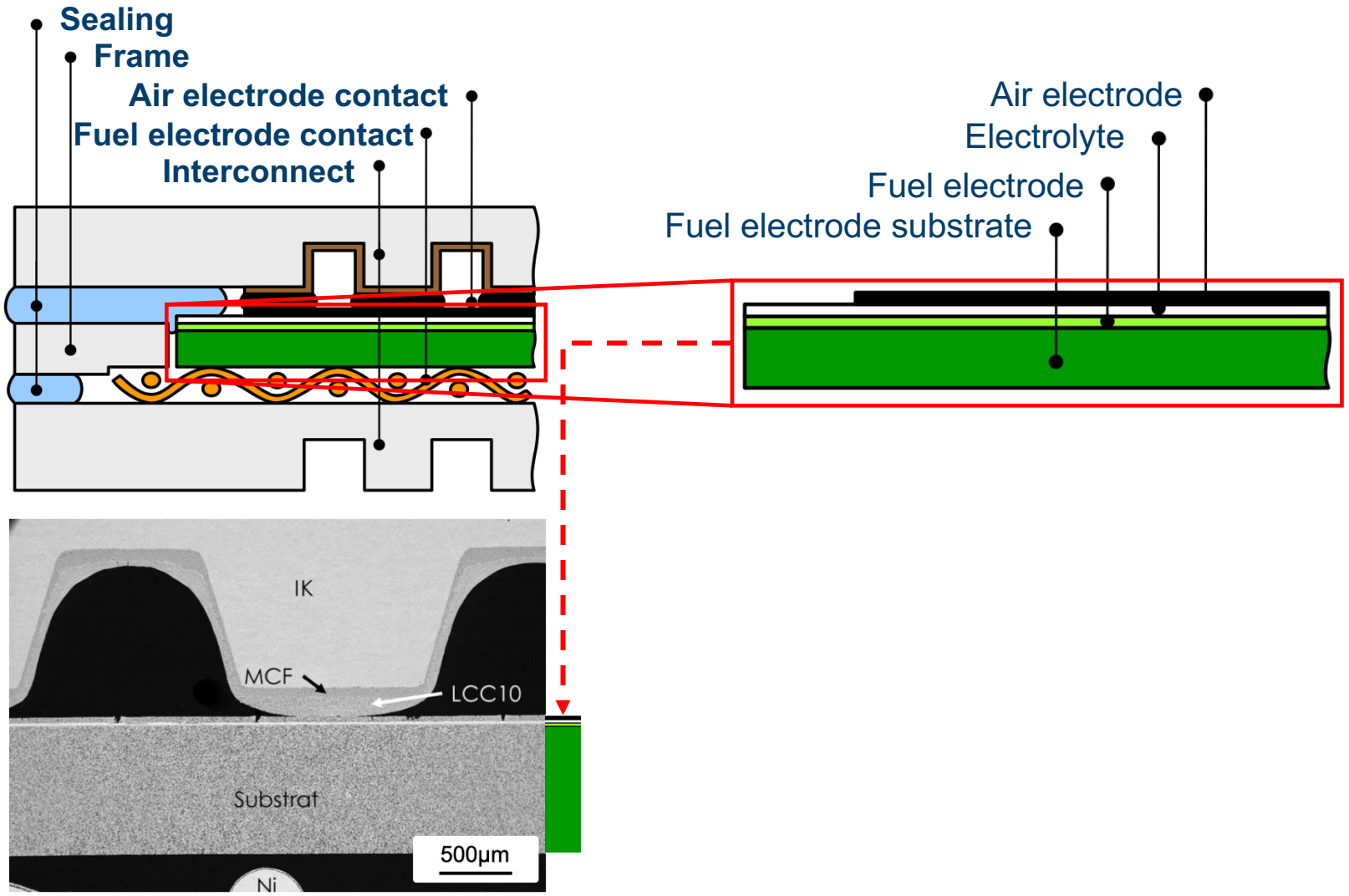

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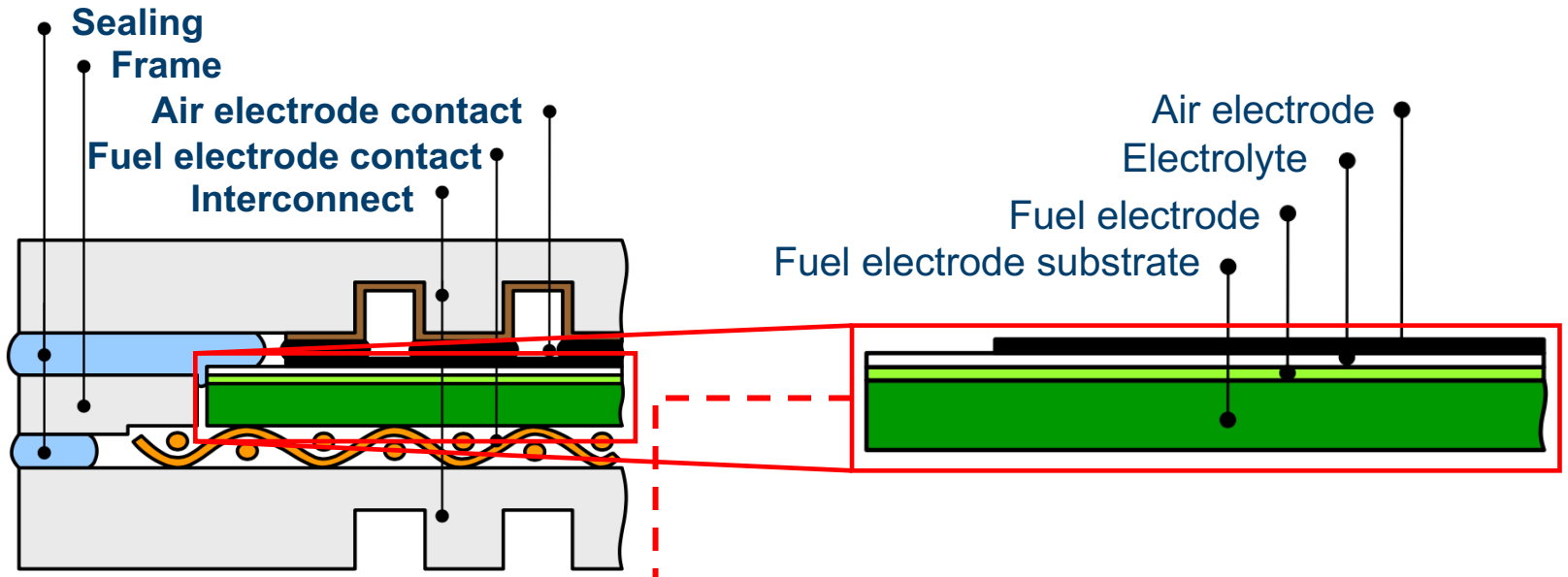

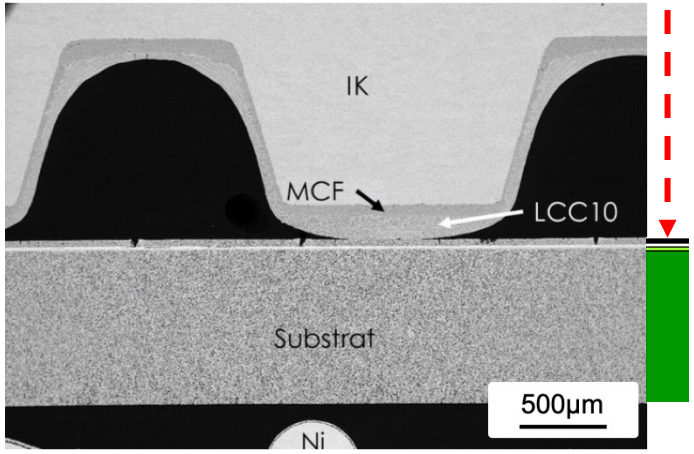

- Support: largest share in cell material
- Requirements: Mech. stabilization, electron conduction, gas transport
- Impact of contaminations potentially weaker than in functional layers

POSSIBLE RECYCLING CONCEPT (FZ JÜLICH)

Step One

Collecting EoL cell fragments

Step Two

Reoxidation of EoL cell fragments

Step Three

Separation: Mechanical or hydrometallurgical

Open-loop raw material recovery

Step Four

Powder preparation: Milling and sieving

Step Five

Integration into manufacturing process

Step Six

Stack testing or stack operation

Recycling concept developed by S. Sarner (IEK-1, Forschungszentrum Jülich)

CELL MANUFACTURING AND RECYCLING LOOPS

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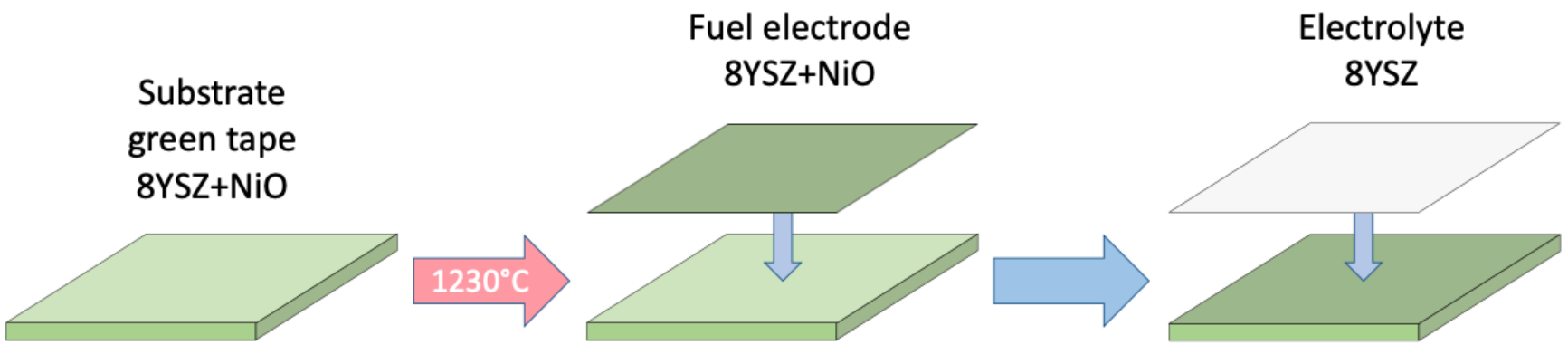

Half cell

8YSZ = 8 mol% Y_2O_3 stabilized ZrO_2

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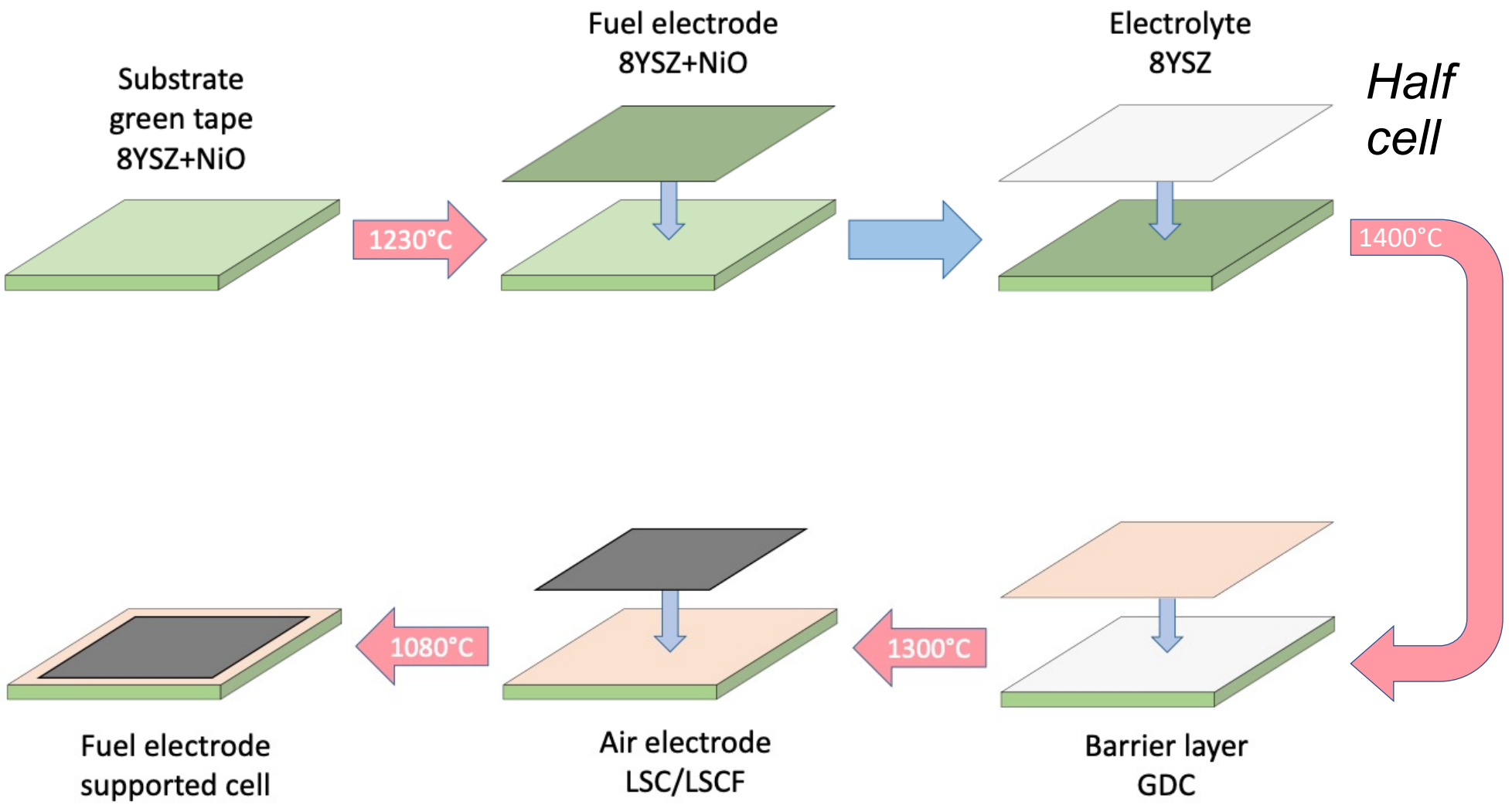

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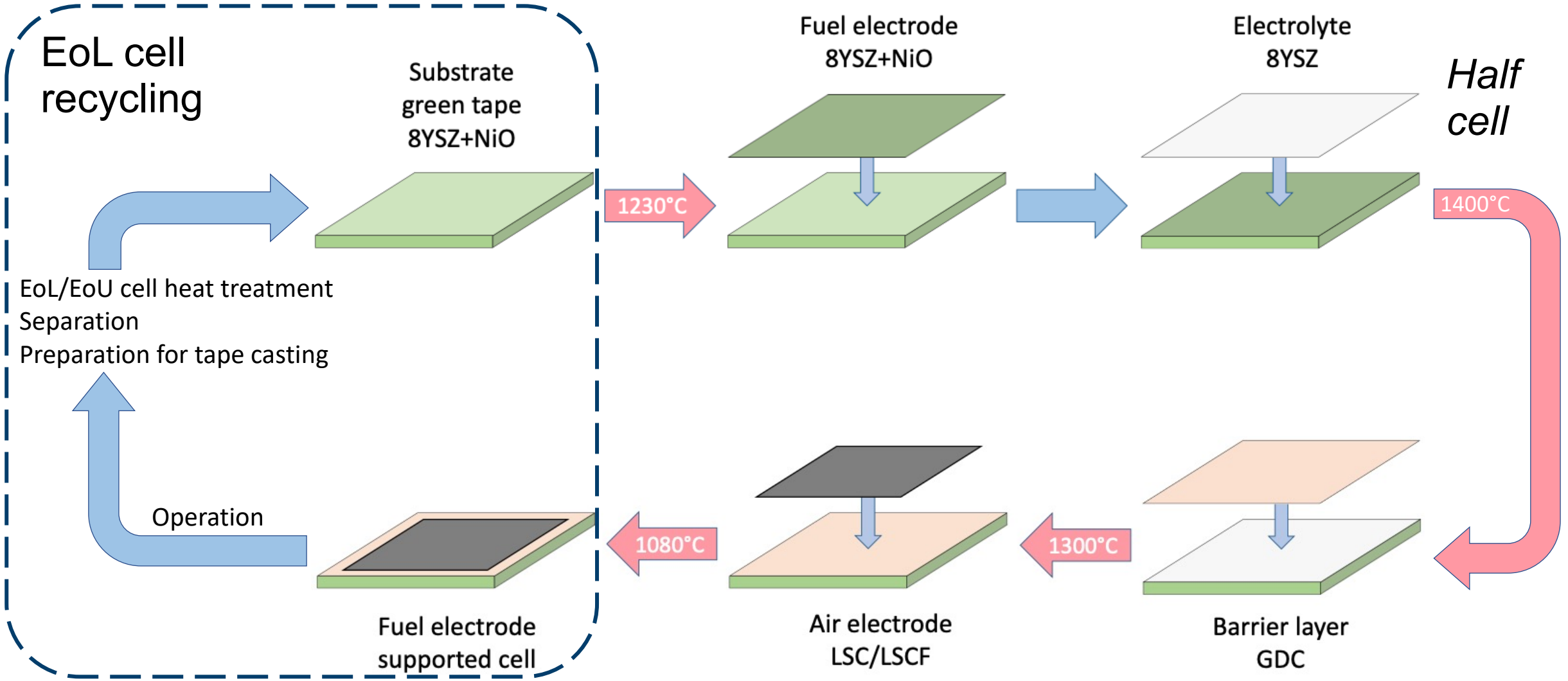

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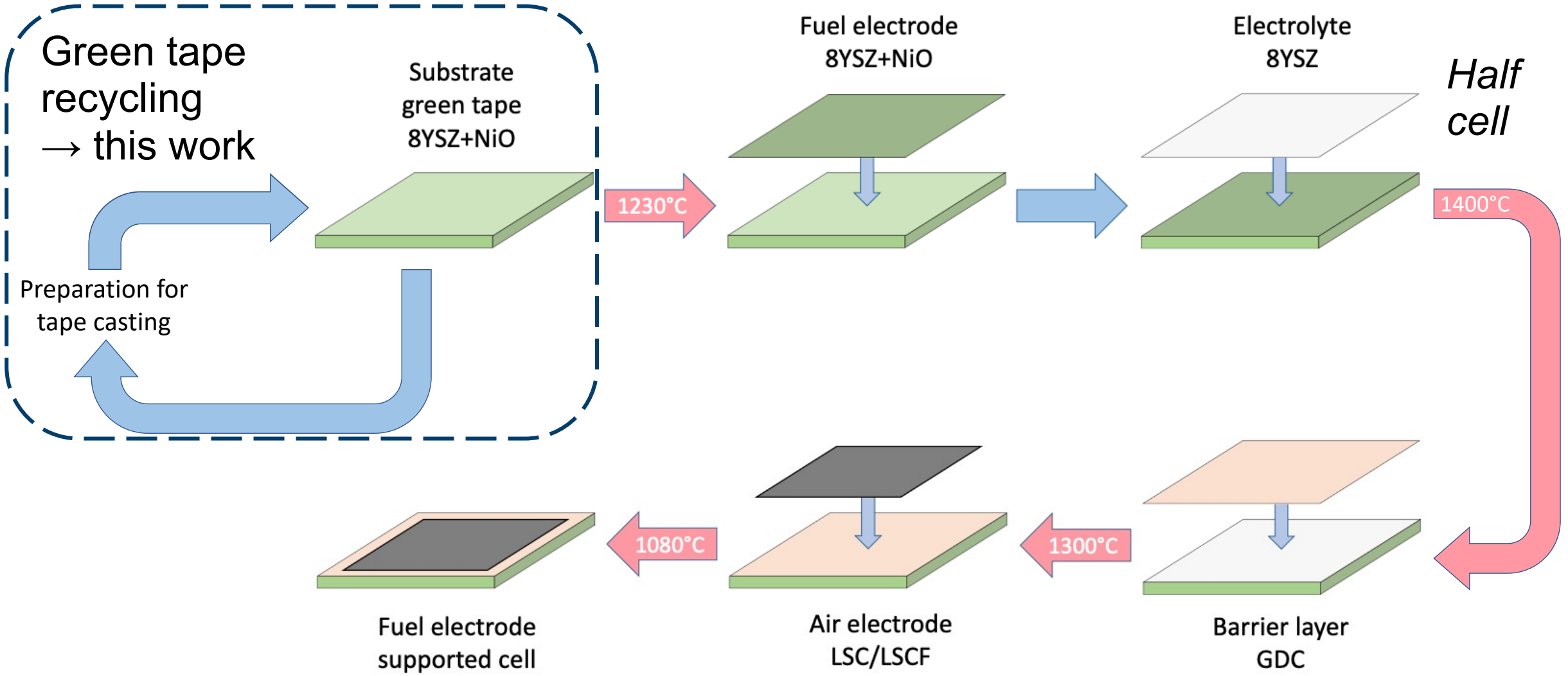

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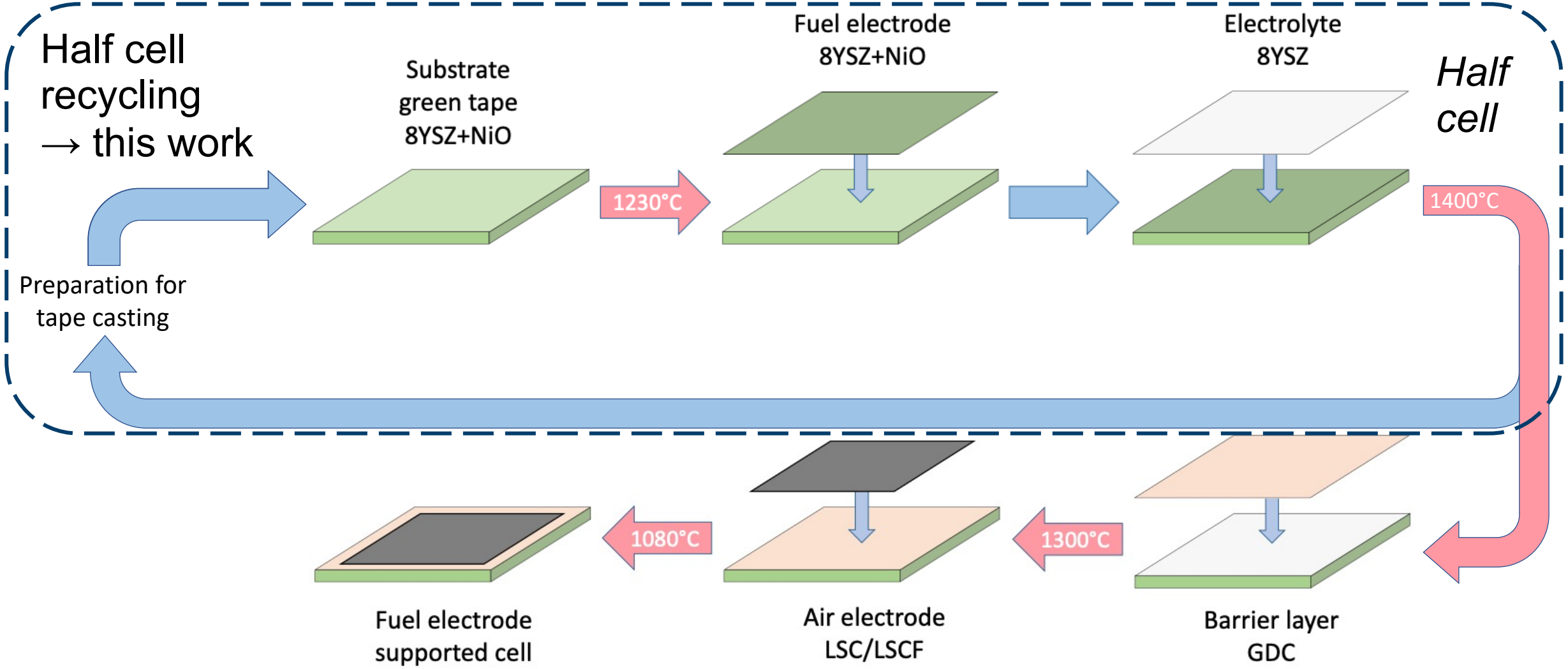

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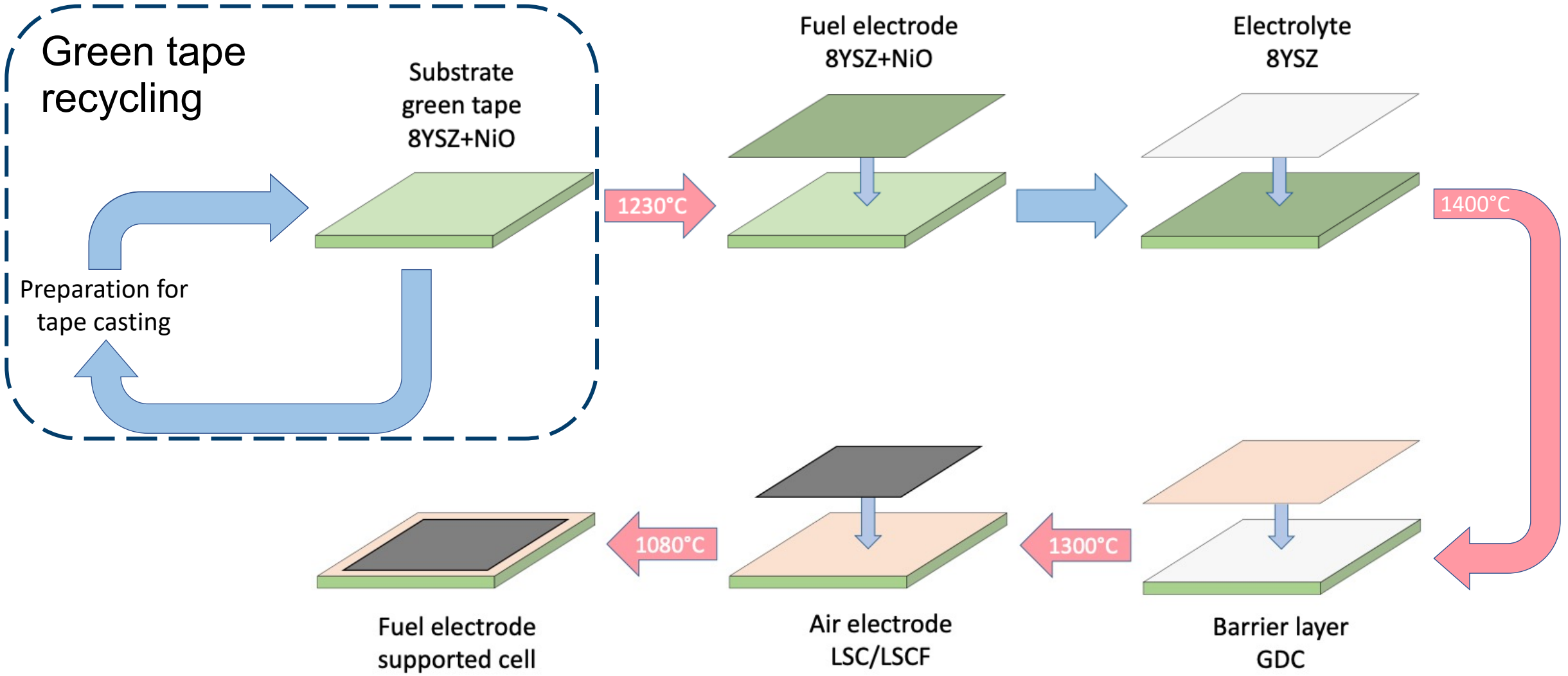
GREEN TAPE RECYCLING



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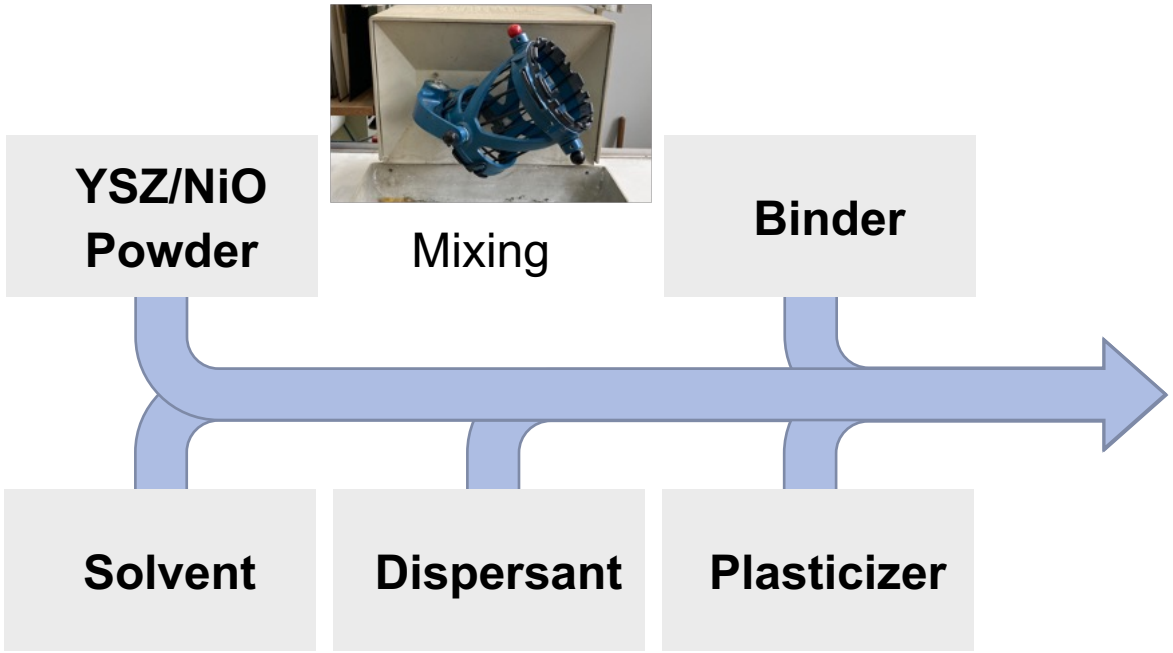
TAPE CASTING AND RECYCLING OF SUBSTRATES



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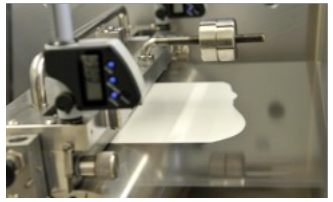

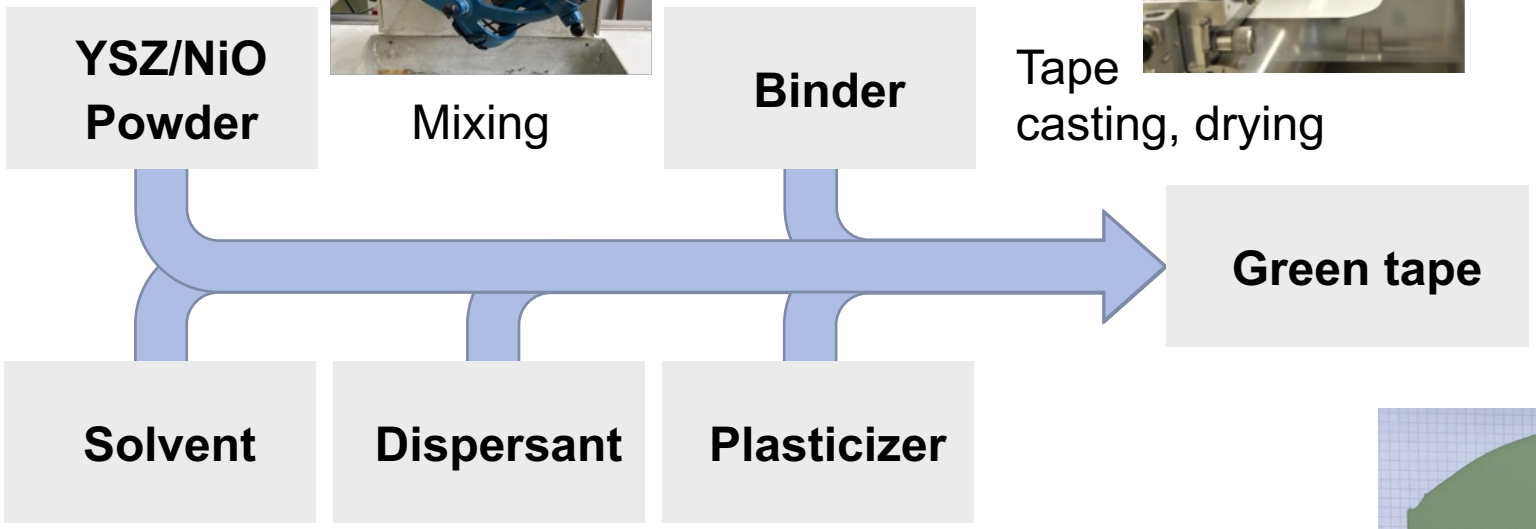

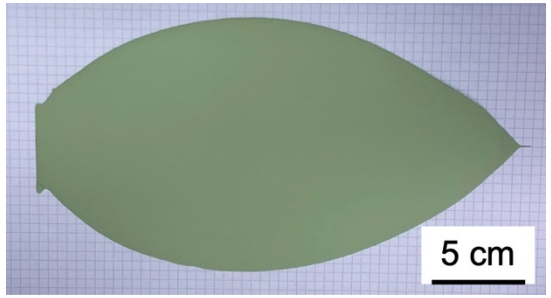

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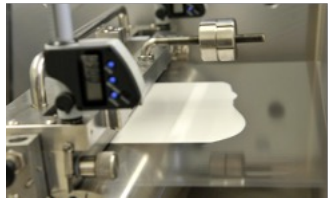


YSZ/NiO Powder

Mixing

Binder

Tape casting, drying

Pre-Sintering

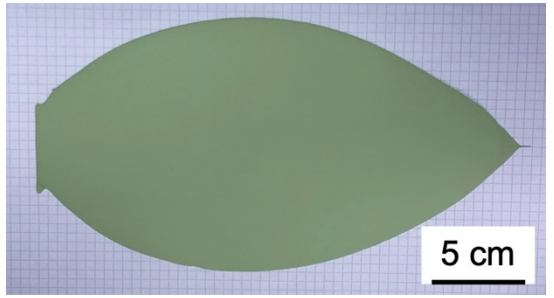
Green tape

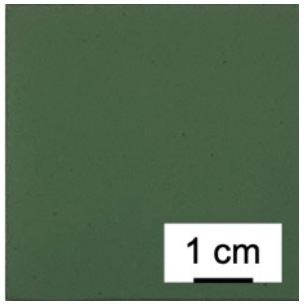
Pre-sintered substrate

Solvent

Dispersant

Plasticizer





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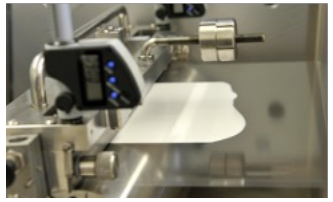


YSZ/NiO Powder

Mixing

Binder

Tape casting, drying

Pre-Sintering

Further processing

Solvent

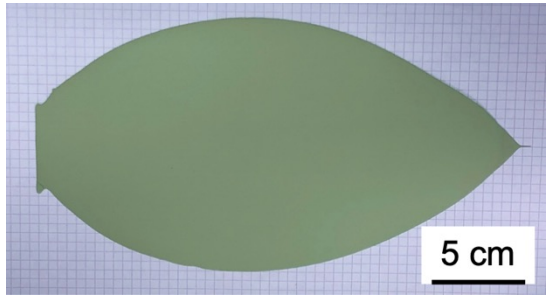
Dispersant

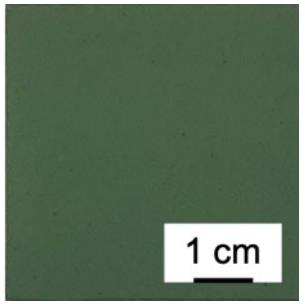
Plasticizer

Green tape

Pre-sintered substrate

…





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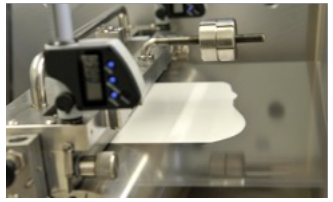

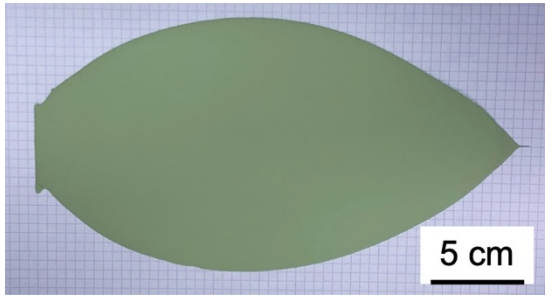

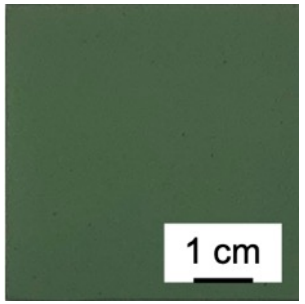

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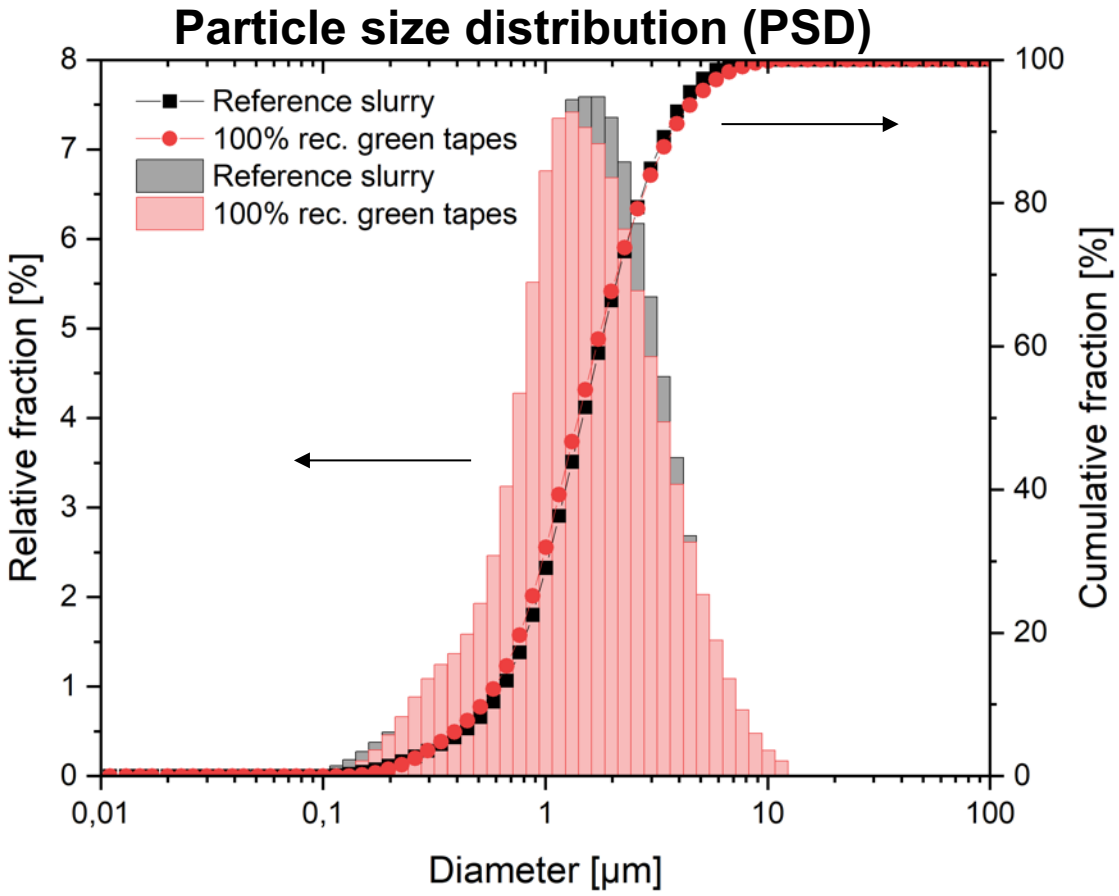

Re-suspending in solvent

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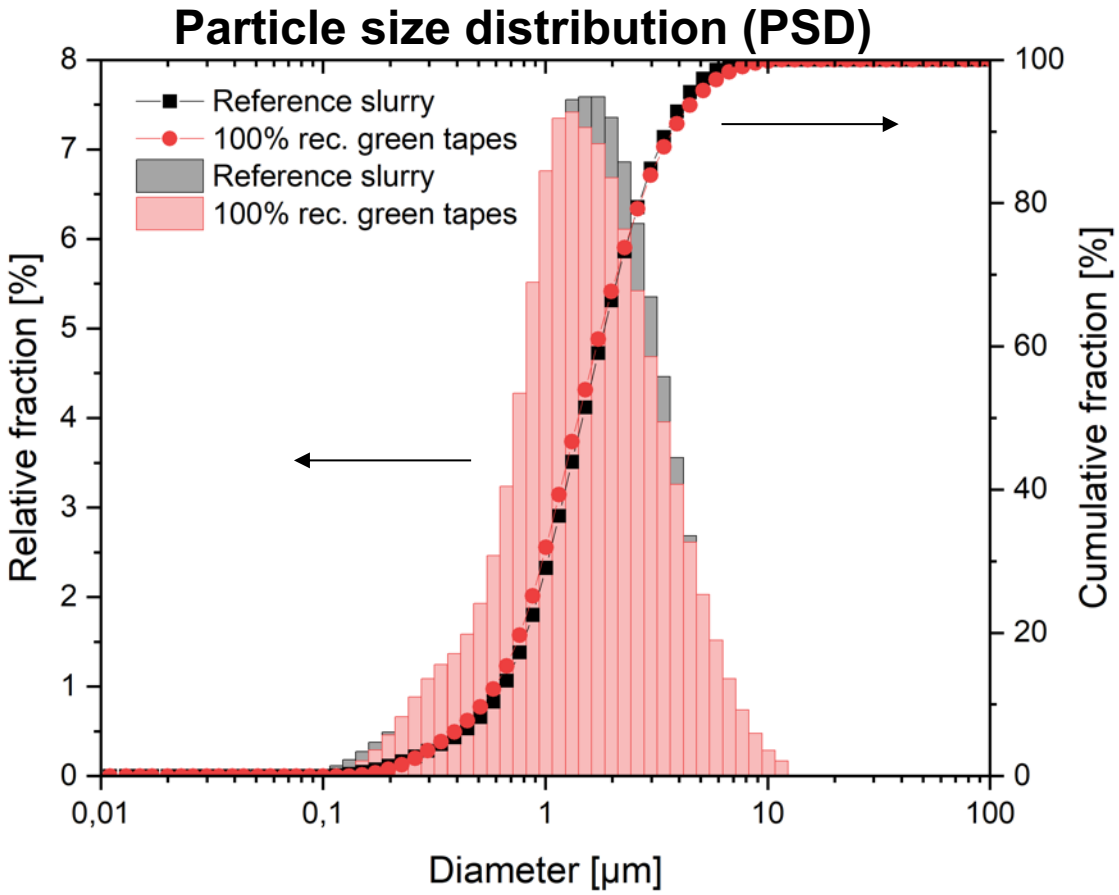

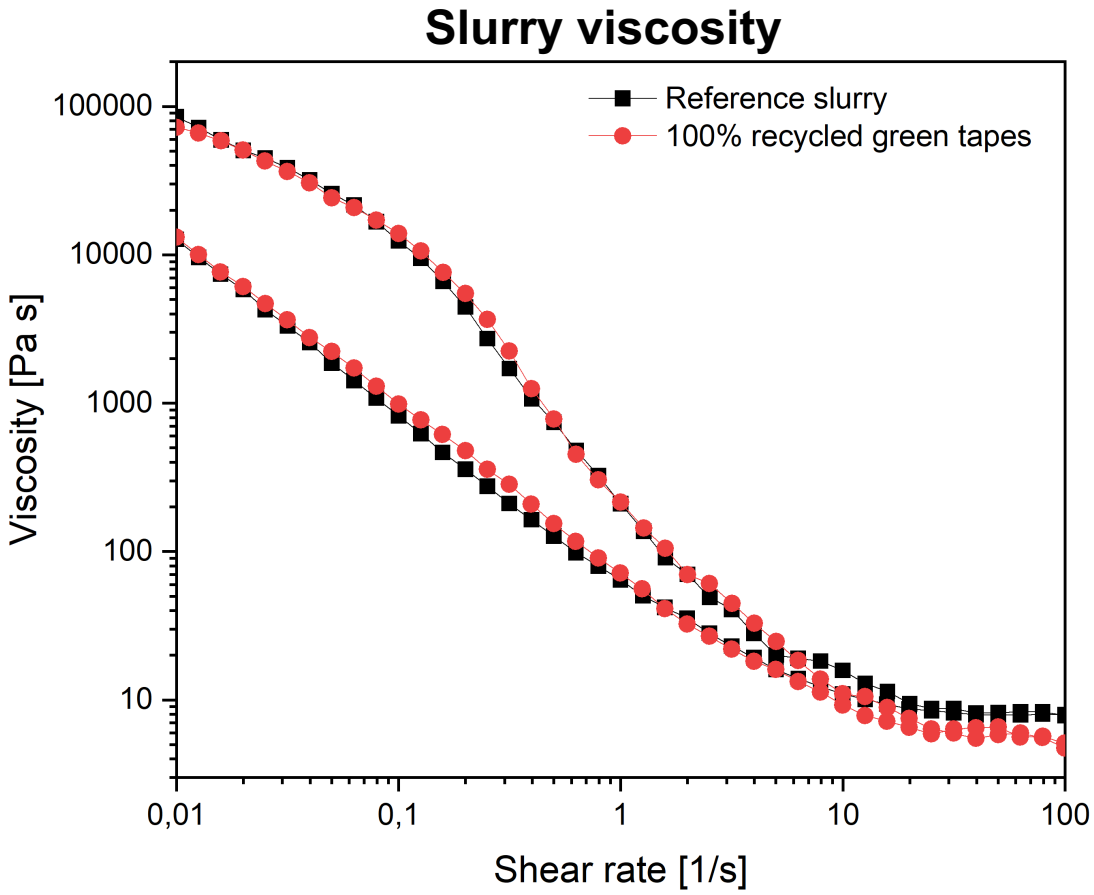

Re-suspending in solvent

Similar slurry properties achieved
Processing via tape casting possible

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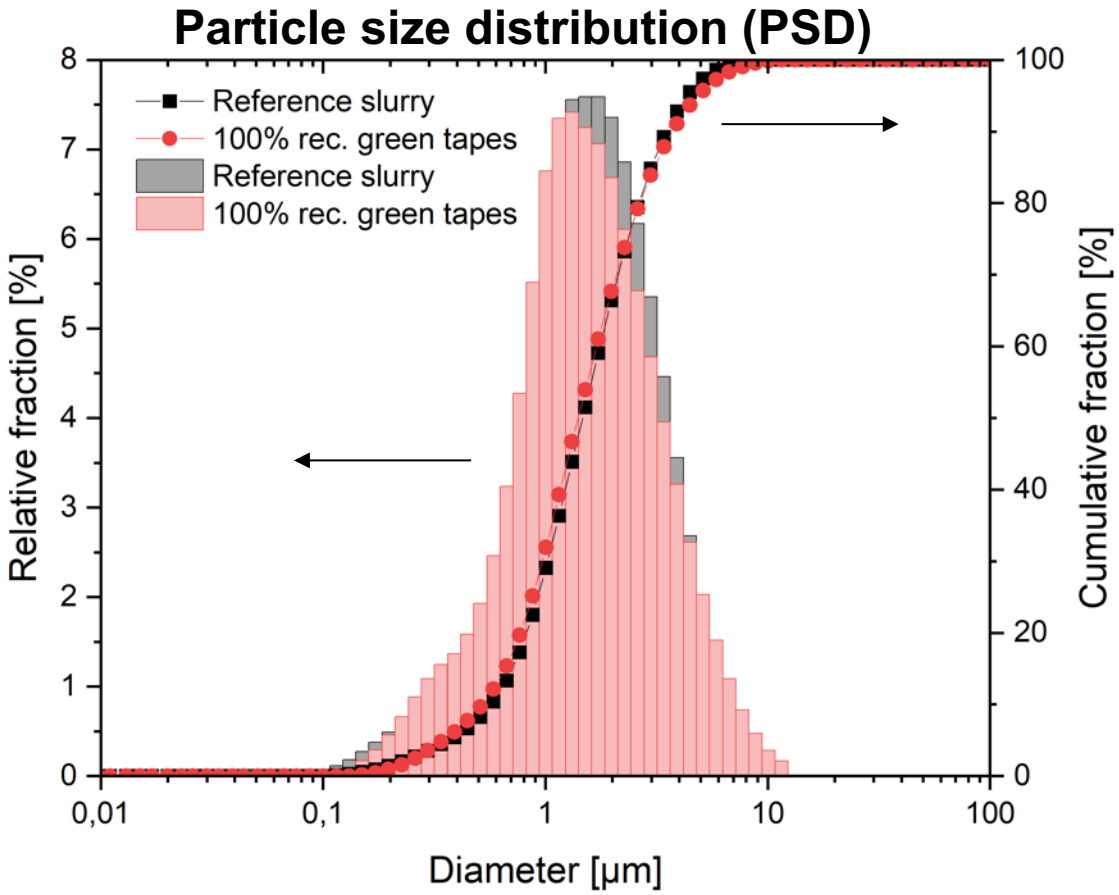

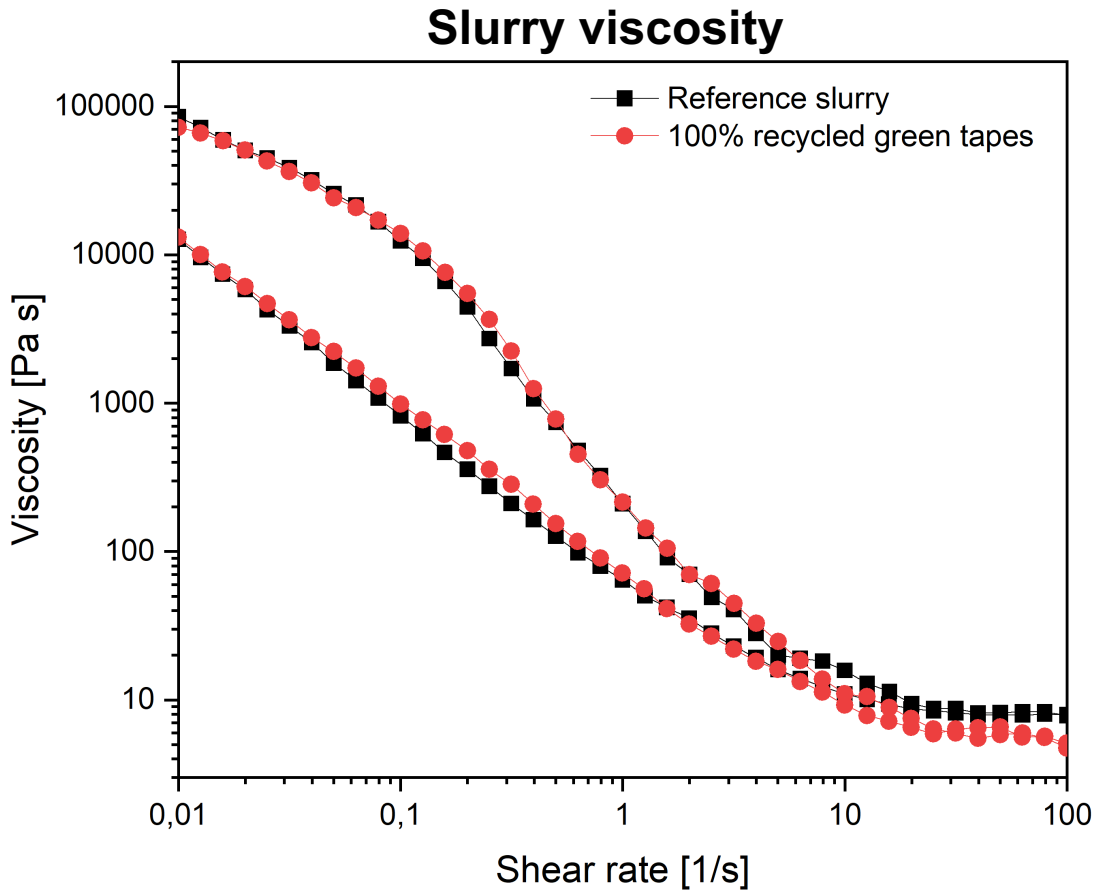

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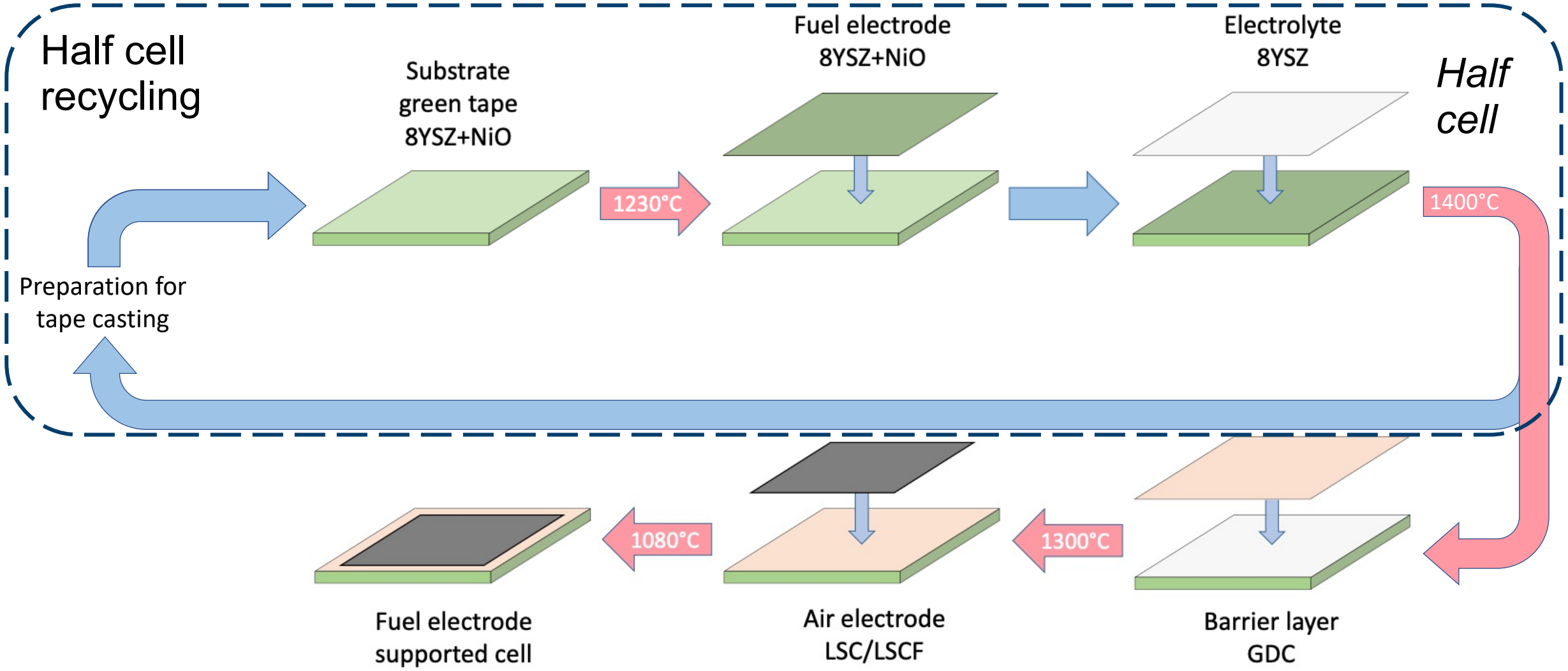
Full recovery possible

RECYCLING OF SINTERED HALF CELLS



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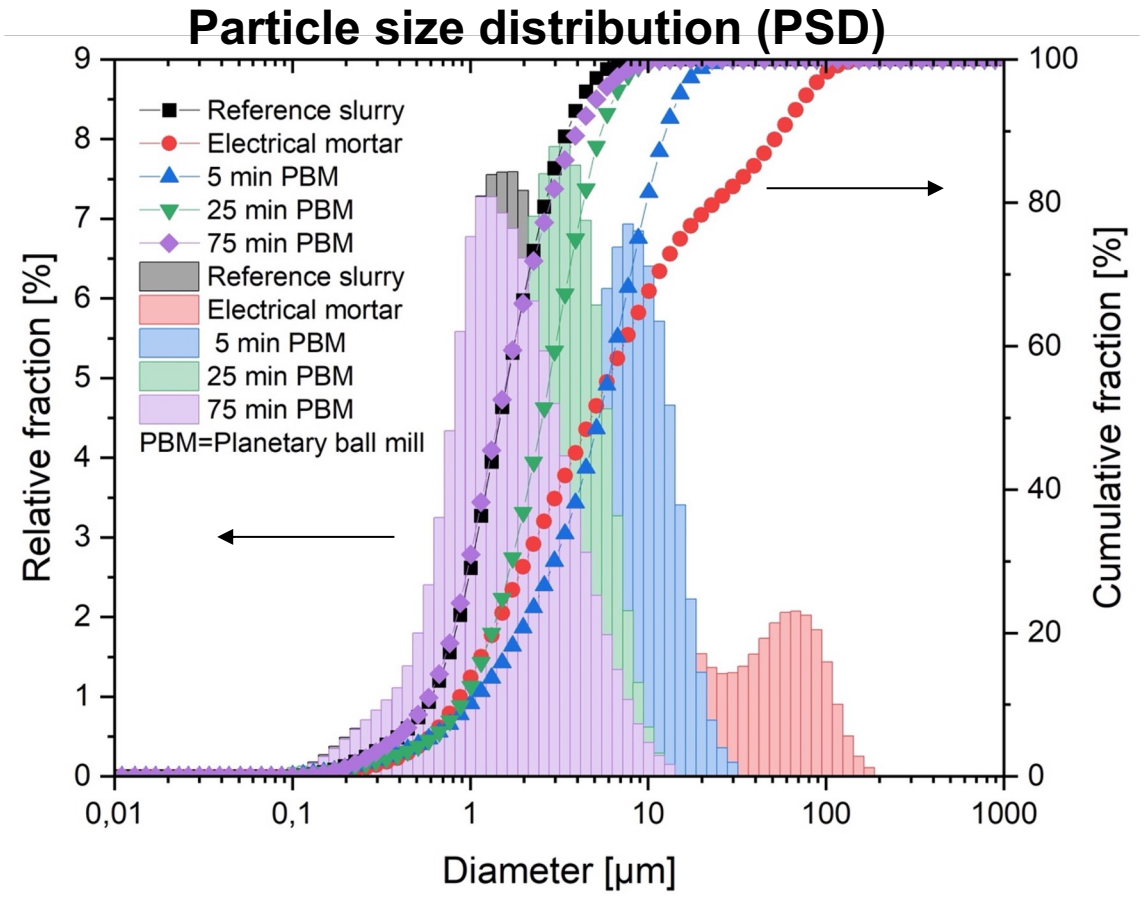
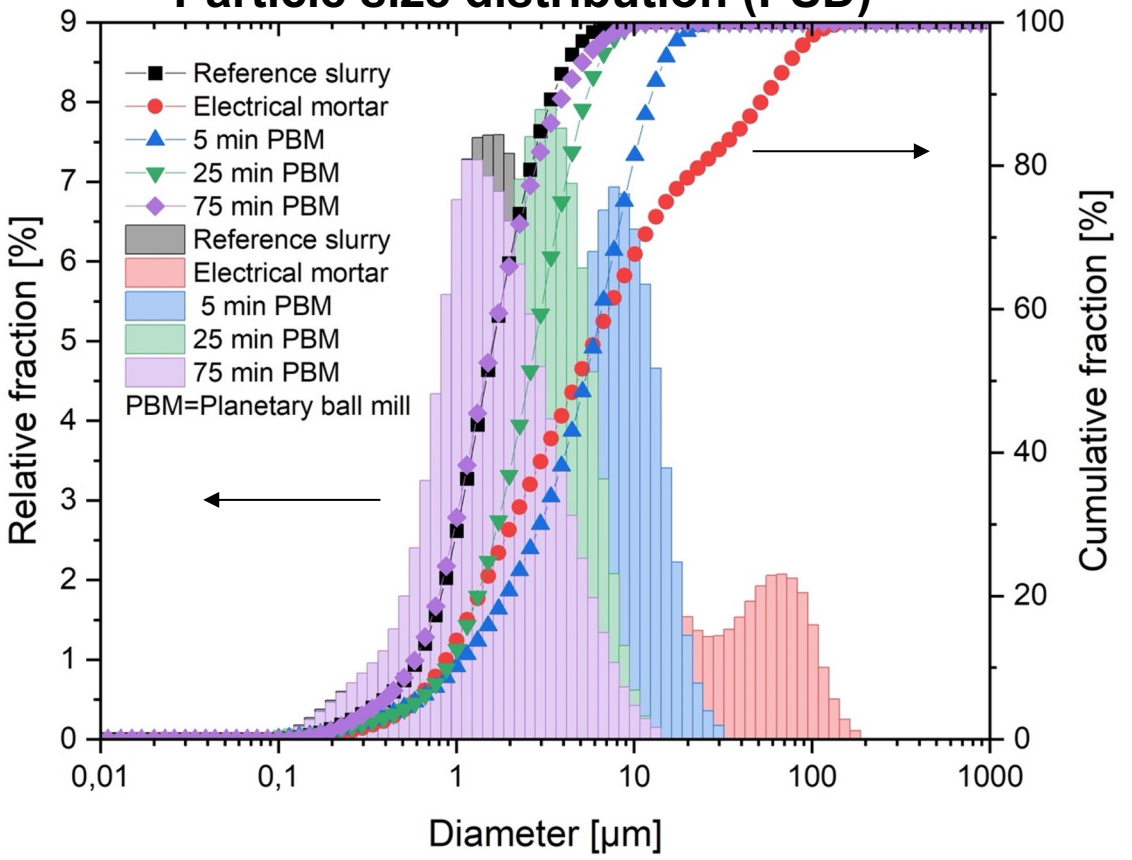
RE-PROCESSING OF SINTERED HALF CELLS



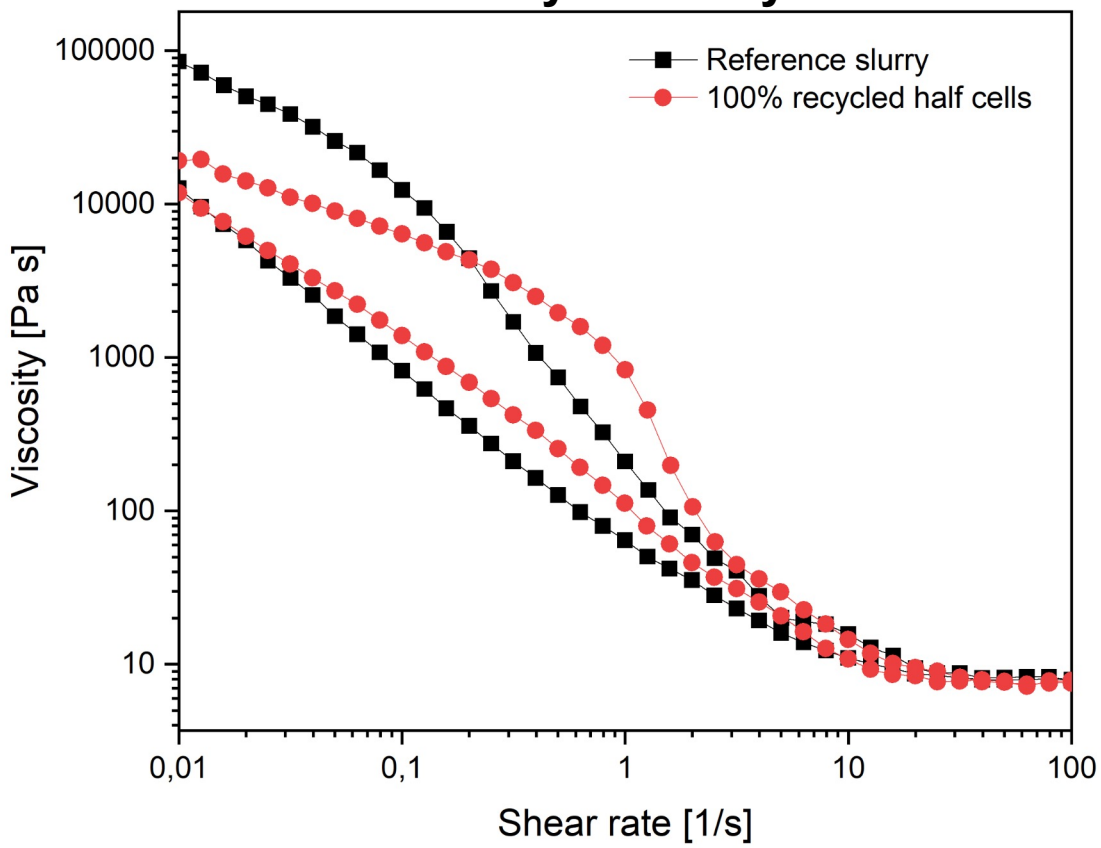
Slurry processing after milling study

RE-PROCESSING OF SINTERED HALF CELLS

Particle size distribution (PSD)



Slurry viscosity



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Influence on rheology

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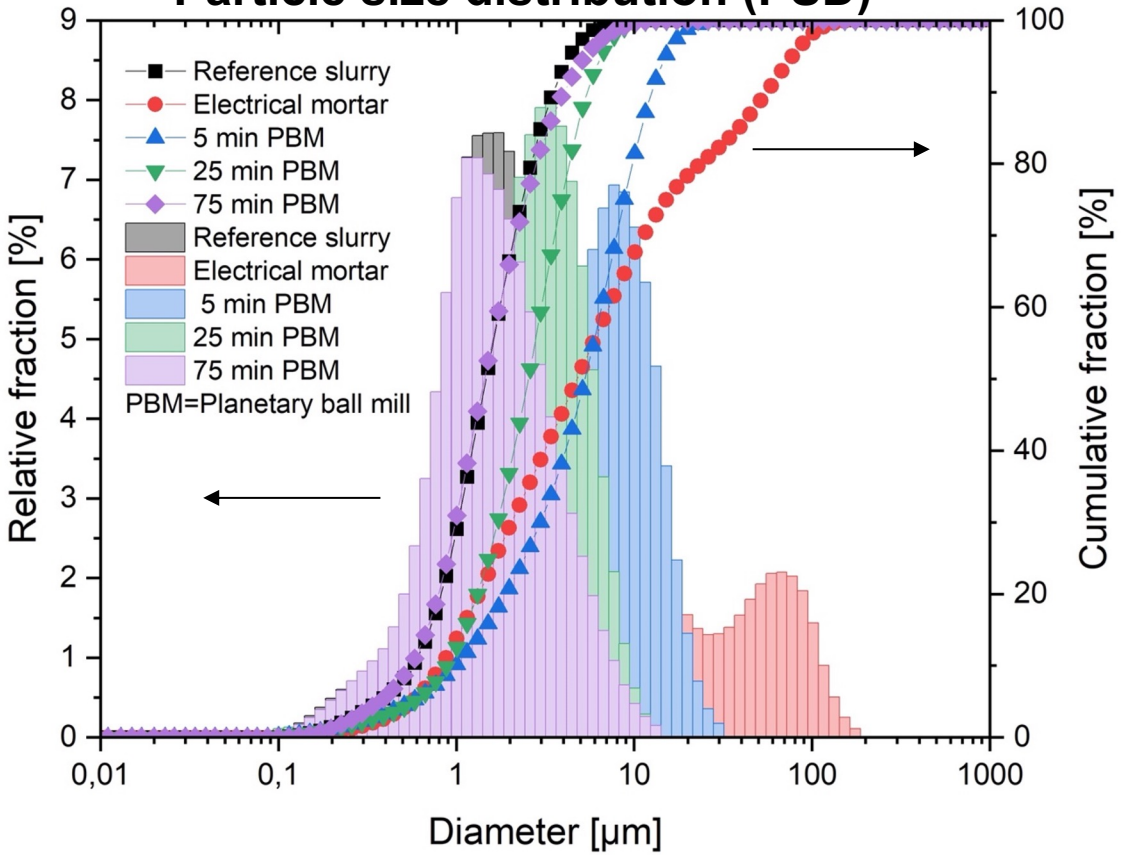

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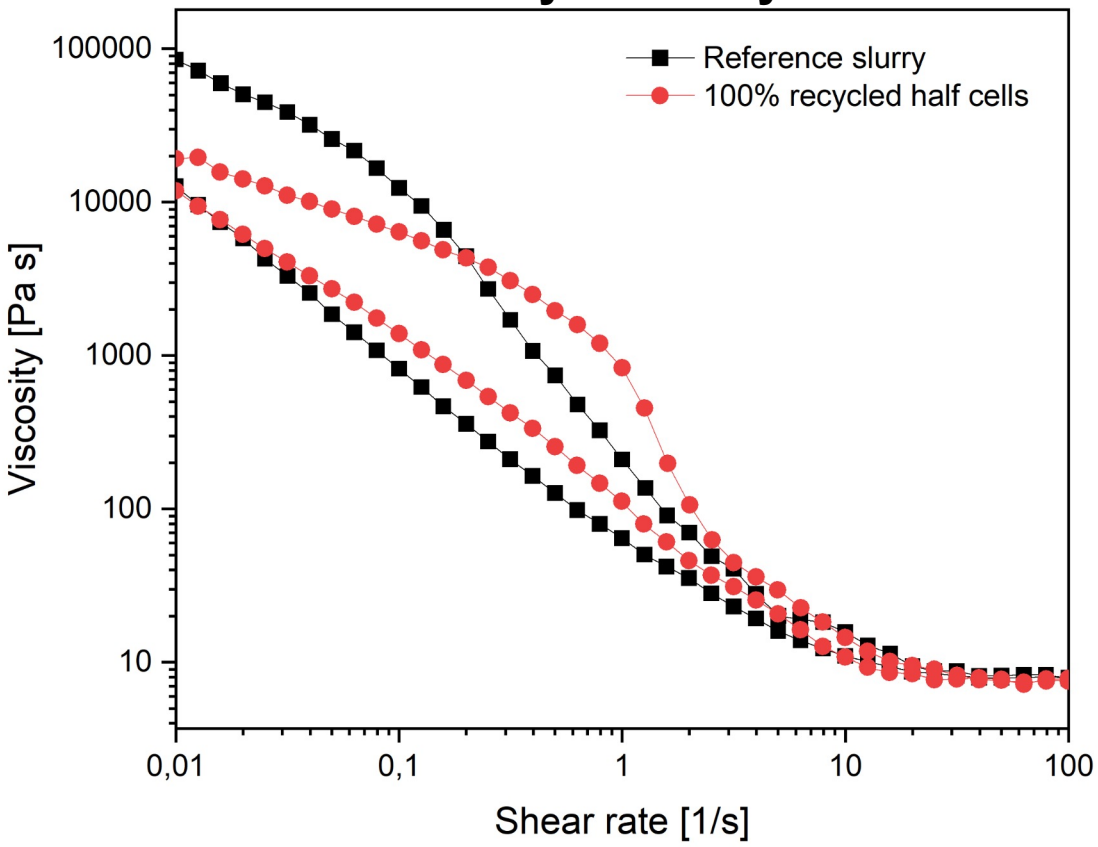

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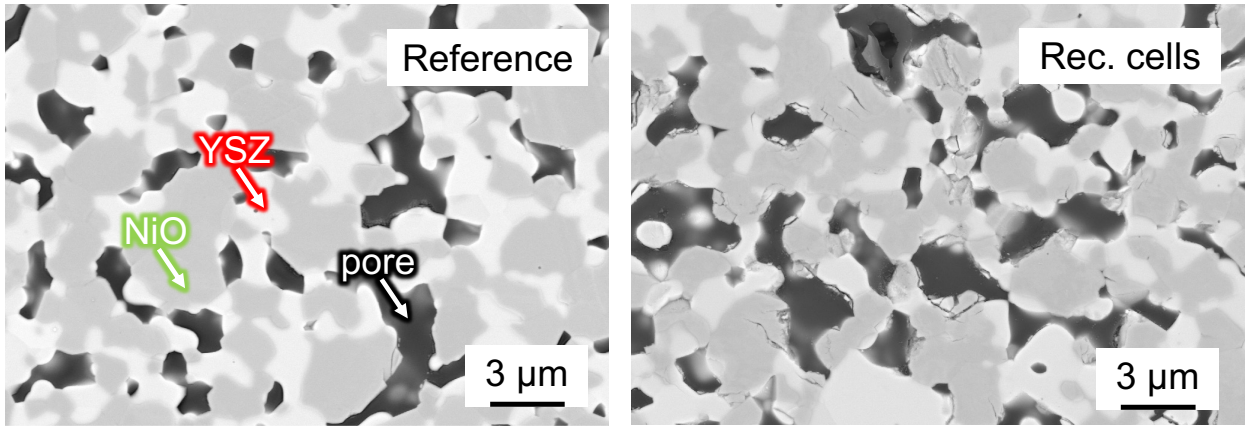
Processing via **tape casting possible**

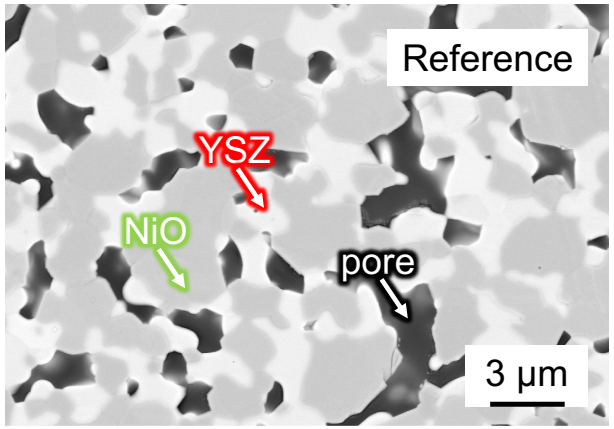
THERMAL PROCESSING OF RECOVERED HALF CELLS

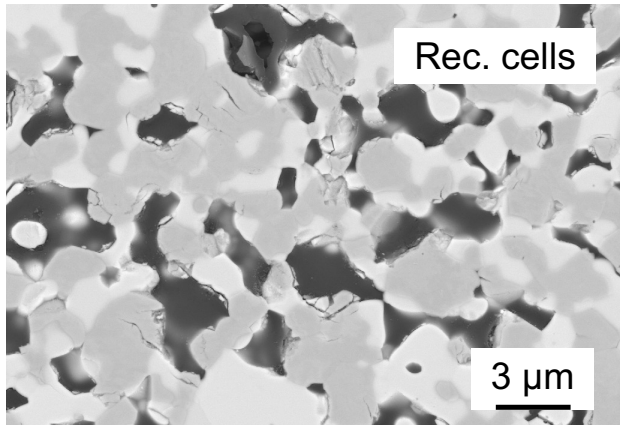
Sintered at 1400°C in air



THERMAL PROCESSING OF RECOVERED HALF CELLS

Sintered at 1400°C in air

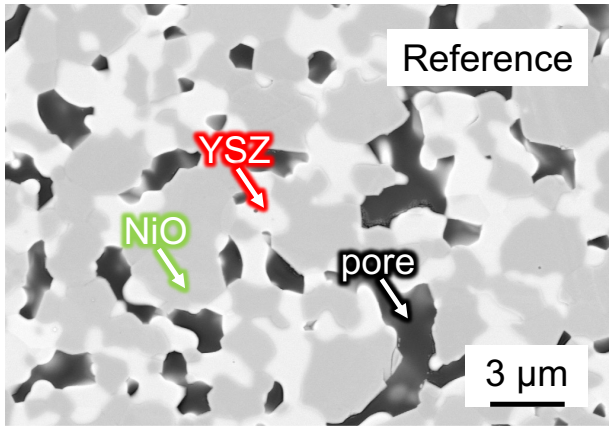


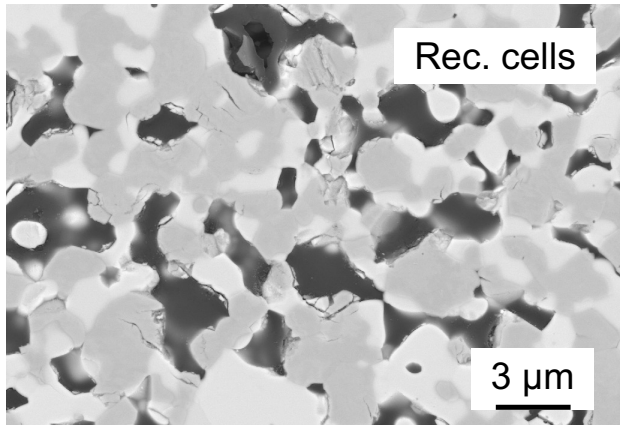


Microstructure	→	Similar pore morphology
Sintering behavior	→	Lowered shrinkage: 13% (Reference 18%)

THERMAL PROCESSING OF RECOVERED HALF CELLS

Sintered at 1400°C in air





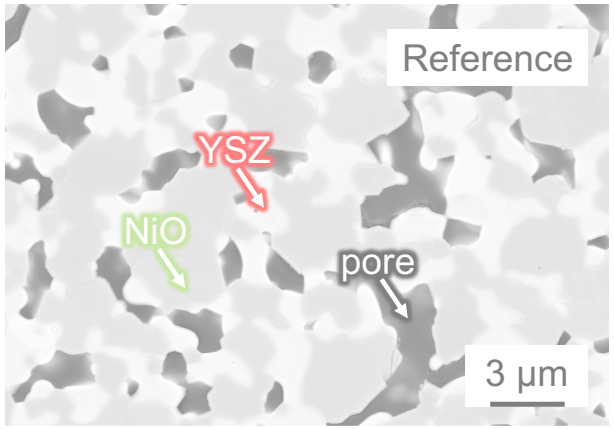
Microstructure	▶	Similar pore morphology
Sintering behavior	▶	Lowered shrinkage: 13% (Reference 18%)

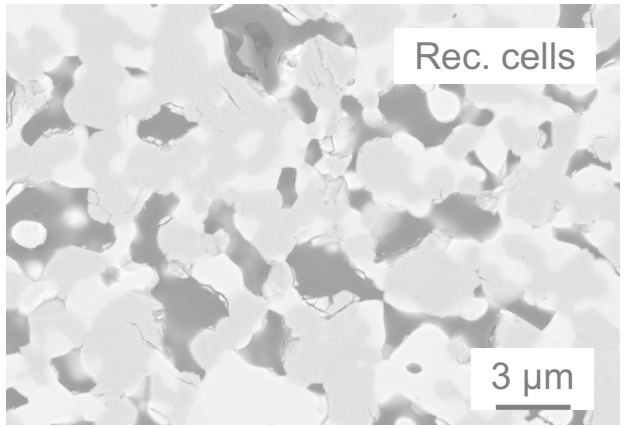
▶ **Affecting further cell processing route**

JÜLICH Forschungszentrum

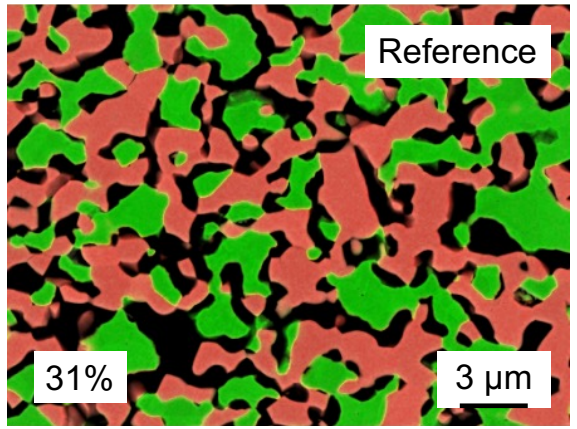
THERMAL PROCESSING OF RECOVERED HALF CELLS

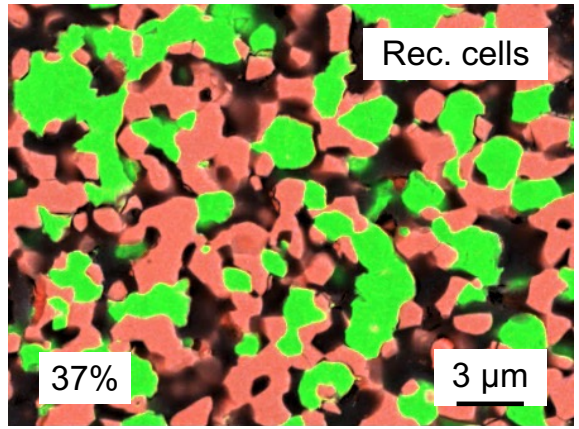
Sintered at 1400°C in air





Reduced at 900°C in H_2/Ar





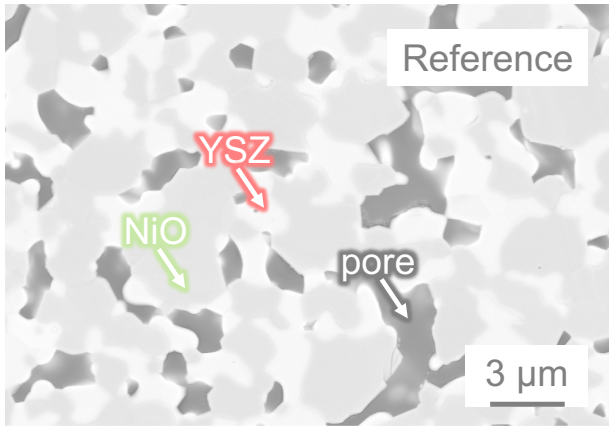
Microstructure	▶	Similar pore morphology
Sintering behavior	▶	Lowered shrinkage: 13% (Reference 18%)

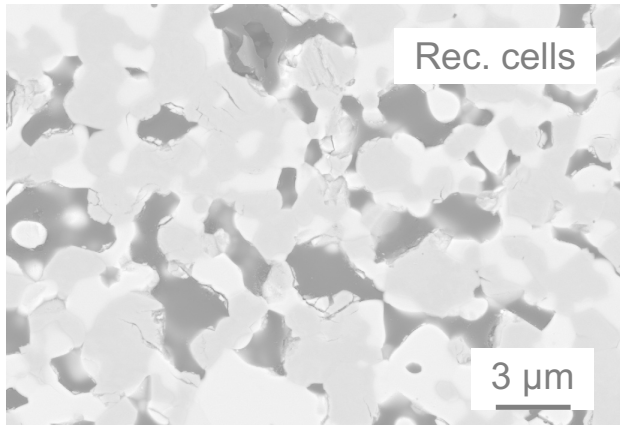
▶ **Affecting further cell processing route**

Green = Ni
Red = Zr

THERMAL PROCESSING OF RECOVERED HALF CELLS

Sintered at 1400°C in air





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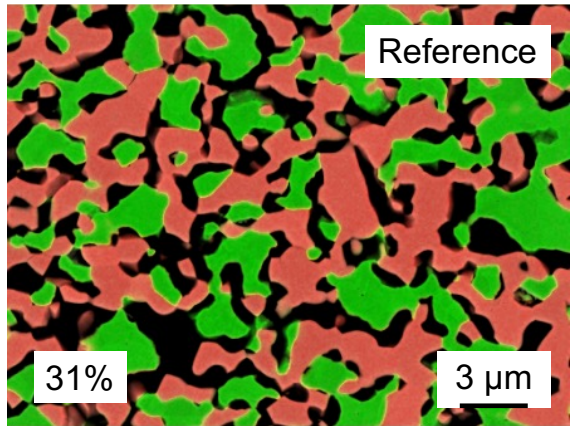

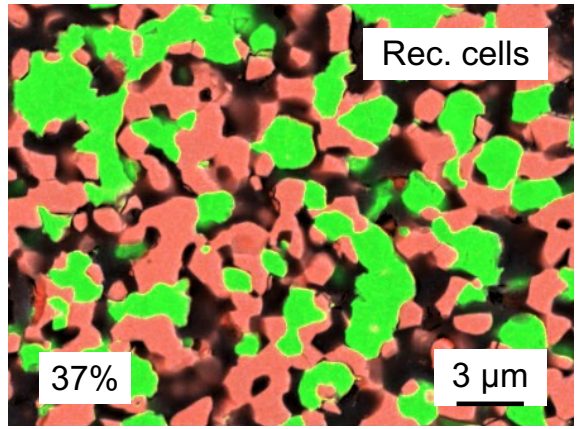

Microstructure	Similar pore morphology	**Microstructure**	Higher porosity Ni grains more spherical
Sintering behavior	Lowered shrinkage: 13% (Reference 18%)	**Performance**	Percolating Ni expected Reduced mech. stability

Affecting further cell processing route

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THERMAL PROCESSING OF RECOVERED HALF CELLS

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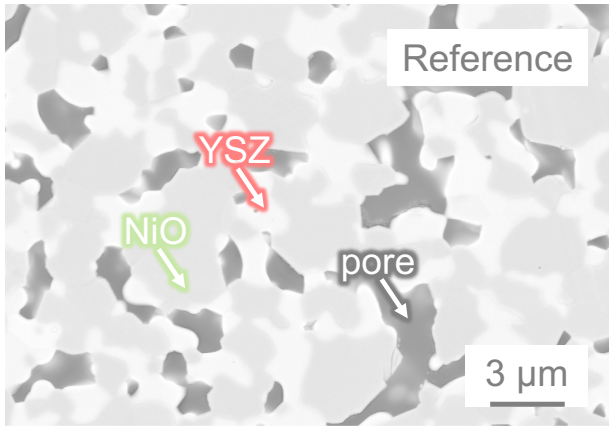

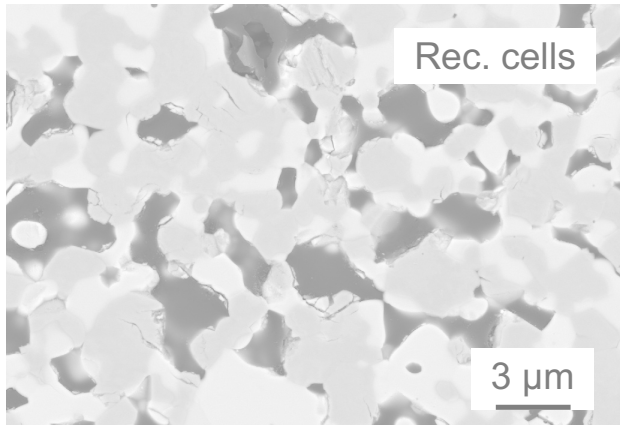

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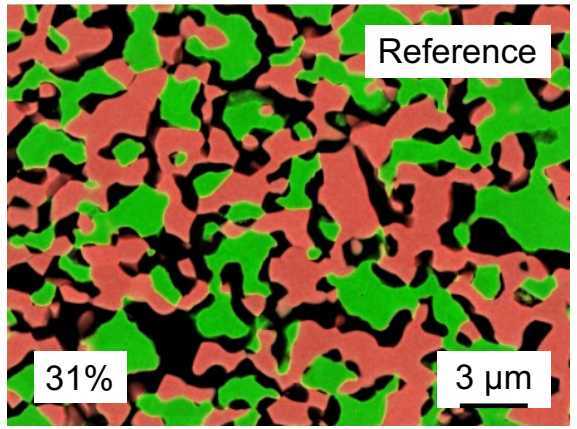

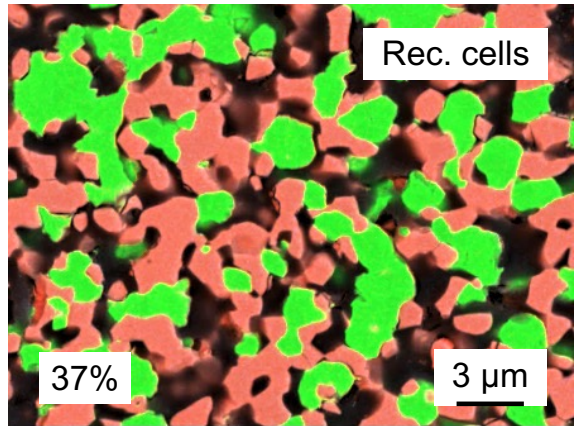

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Sintering behavior	Lowered shrinkage: 13% (Reference 18%)	**Performance**	Percolating Ni expected Reduced mech. stability

Affecting further cell processing route

100% recycling challenging

Green = Ni
Red = Zr

CONCLUSION

Recycling of substrate green tapes

- Re-suspending green tapes to slurry ✓
- Nearly no influence on slurry properties ✓
- **Full recovery of substrate in process** ✓

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CONCLUSION

Recycling of substrate green tapes

- Re-suspending green tapes to slurry ✓
- Nearly no influence on slurry properties ✓
- **Full recovery of substrate in process** ✓
- **Promising to be implemented into manufacturing of SOCs** ☺

Recycling of sintered half cell material

- Processing slurry from re-milled half cells ✓

CONCLUSION

Recycling of substrate green tapes

- Re-suspending green tapes to slurry ✓
- Nearly no influence on slurry properties ✓
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Promising to be implemented into manufacturing of SOCs ☺

Recycling of sintered half cell material

- Processing slurry from re-milled half cells ✓
- Slurry properties, shrinkage, microstructure and mechanical properties affected —
- **Full recovery of sintered cells difficult** —

CONCLUSION

Recycling of substrate green tapes

- Re-suspending green tapes to slurry ✓
- Nearly no influence on slurry properties ✓
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- **Promising to be implemented into manufacturing of SOCs** ☺

Recycling of sintered half cell material

- Processing slurry from re-milled half cells ✓
- Slurry properties, shrinkage, microstructure and mechanical properties affected —
- **Full recovery of sintered cells difficult** —
- **Recycling of defined shares realistic Adaptability for End-of-Life material?** ☺

Currently investigated at IEK-1

THANK YOU FOR YOUR ATTENTION!

Special thanks to:

Prof. Dr. Olivier Guillon	Head of IEK-1
Prof. Dr. Jesus Gonzalez-Julian	First examinor
Hon.-Prof. Dr. Norbert H. Menzler	Second examinor
Stephan Sarner, M.Sc.	Scientific advisor
Ralf Kauert	Slurry preparation
Andrea Hilgers	PSD analysis, Rheology
Dr. Doris Sebold	SEM, EDS
Volker Bader	Heat treatments