



Membrane-assisted Ethylene Synthesis over
Nanostructured Tandem Catalysts

CO₂ conversion to green ethylene: a tandem CZA-zeolite catalyst approach

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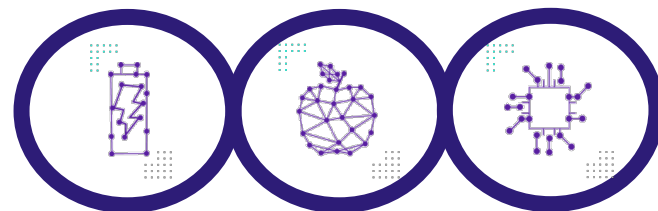
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Energy and
Environment

Life
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Technologies



KOLEN'KO RESEARCH GROUP

Where Transport Meets Reaction

- Heterogeneous catalysis
 - Electrolysis
 - Coatings





MemCat Project



Membrane-assisted Ethylene Synthesis over Nanostructured Tandem Catalysts

Why Ethylene?

1.2 tons
of CO₂

per ton of produced ethylene!

Mackenzie, 09 April 2024.



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Current production & utilization of ethylene



Diverse Applications of Ethylene



Healthcare Applications: Ethylene derivatives are essential in manufacturing medical devices, sterilization agents, and pharmaceuticals



Renewable Energy Materials: Ethylene-based materials contribute to solar panel production and wind turbine components



Advanced Industrial Materials: Ethylene is vital in developing advanced materials for construction, automotive, and electronics industries



MemCat Project

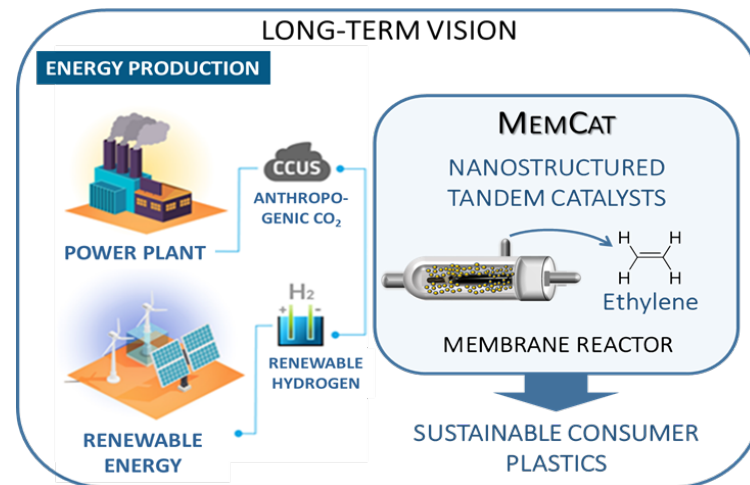


Membrane-assisted Ethylene Synthesis over Nanostructured Tandem Catalysts

The goal of MemCat:

Deliver a proof-of-concept for carbon dioxide (CO₂) direct conversion to ethylene (ET)

using tandem catalysts within a membrane reactor



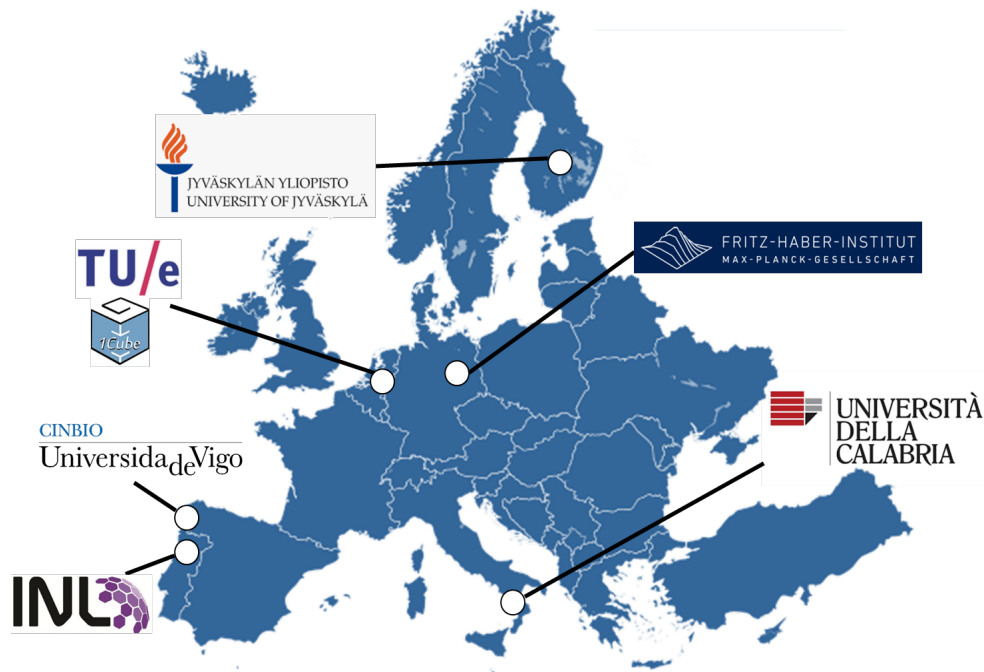
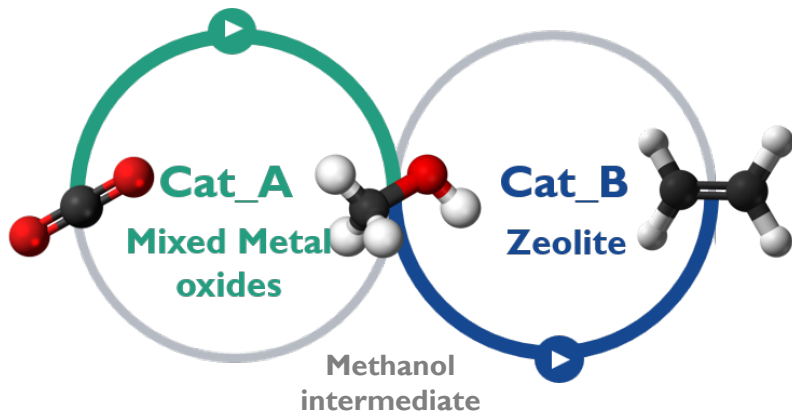
❖ CO₂ hydrogenation to Ethylene with $X \geq 80\%$ and $S \geq 95\%$



MemCat Project



Our role



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CO₂ to light olefins



Four matching rules for constructing high-performance bifunctional MCs-zeolites-based catalysts

Catalysis Reviews, 2024, 1-76

1

Methanol catalyst:

High CO₂-to-methanol selectivity under high temperatures

2

Zeolite:

Appropriated topology, moderate acidity, and pore-structure to high selectivity

3

Efficient matching in conversion rate:

MeOH formation higher than its conversion rate: unconverted MeOH can be decomposed to CO;

MeOH formation lower: zeolite-efficiency cannot be fully utilized, and the over-hydrogenation will form paraffin

4

Unique spatial distributions:

Too far, diffusion distance of MeOH prolonged: concentration gradient and decreases the catalytic activity;

Too close, active sites can poison each other: loss of catalytic activity.

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Catalyst synthesis



Tandem Catalyst Materials selection

Cat_ A_{MeOH} : Metal oxides



Co-precipitation procedure

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MAX-PLANCK-GESELLSCHAFT



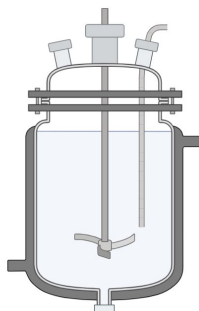
Mixed Metal oxides

500 mL of nitrates



+

Base (pH control) or
precipitation agent



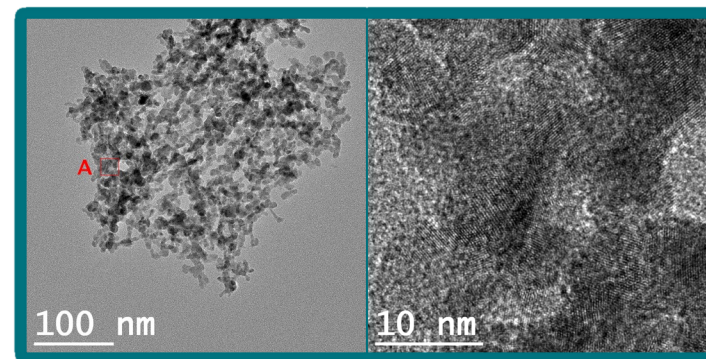
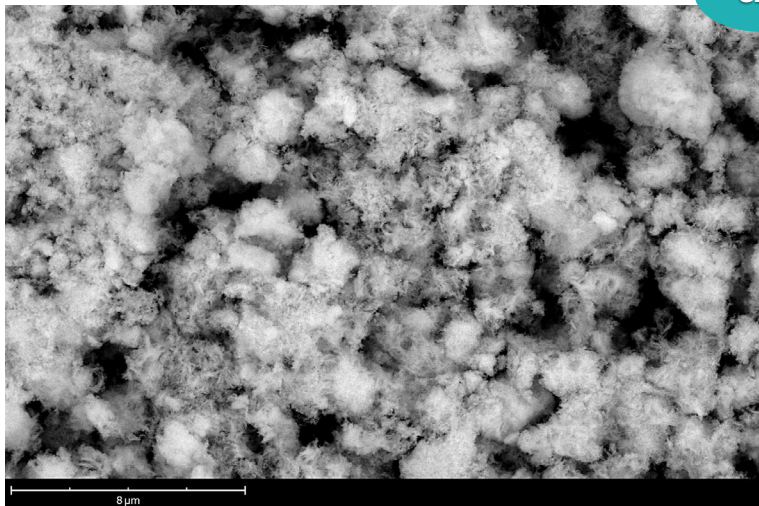
MQ water



Tandem Catalyst Materials selection

Morphology

CZA



Crystallite size: 9 nm ± 1 nm

	Cu (%)	Zn (%)	Al (%)	O (%)	Cu	Zn	Al
XRF data	31.7	12.5	0.8	55.0	42.5	16.8	1.0

$S_{\text{BET}} = 90.93 \text{ m}^2/\text{g}$
 $V_{\text{pore}} = 0.524 \text{ cm}^3/\text{g}$
 $D_p = 8.1 \text{ nm}$

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Preliminary results



Tandem Catalyst Materials selection

Catalytic activity



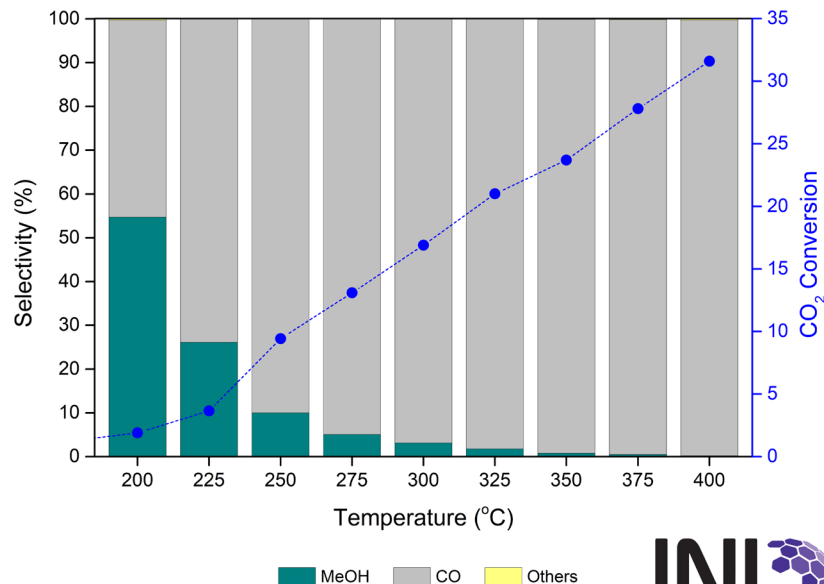
FRITZ-HABER-INSTITUT
MAX-PLANCK-GESELLSCHAFT

K. Kappis, M. Borrelli, A. Trunschke

WHSV: 16800 ml/g/h

$\text{CO}_2 : \text{H}_2 \rightarrow 1:3$

CZA



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Catalyst synthesis

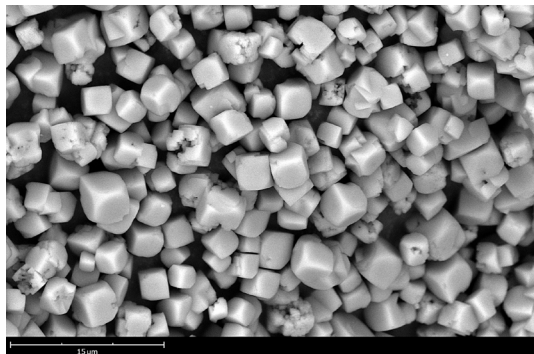


Tandem Catalyst Materials selection

Basic characterization

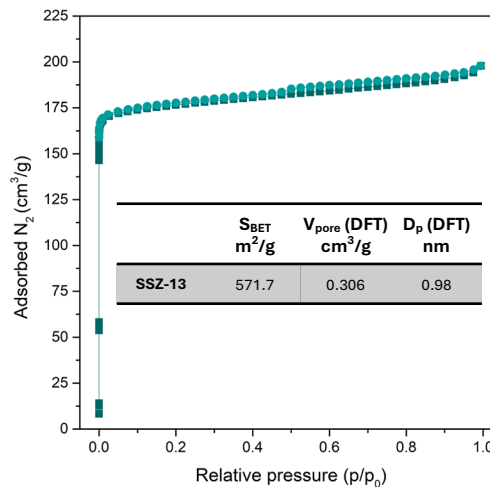
Cat_B_{MTO}: SSZ-13 Zeolite
Commercially available
TOSOH CORPORATION

Morphology



Particle size:
2.7 $\mu\text{m} \pm 0.4$

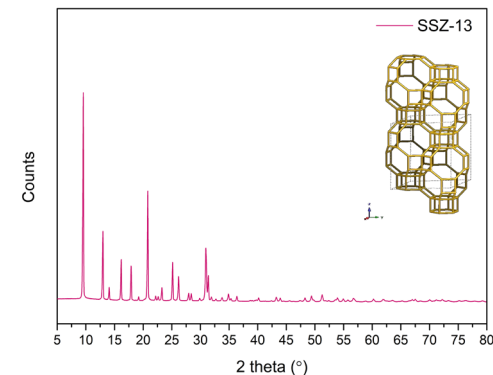
Porosity



Elemental composition

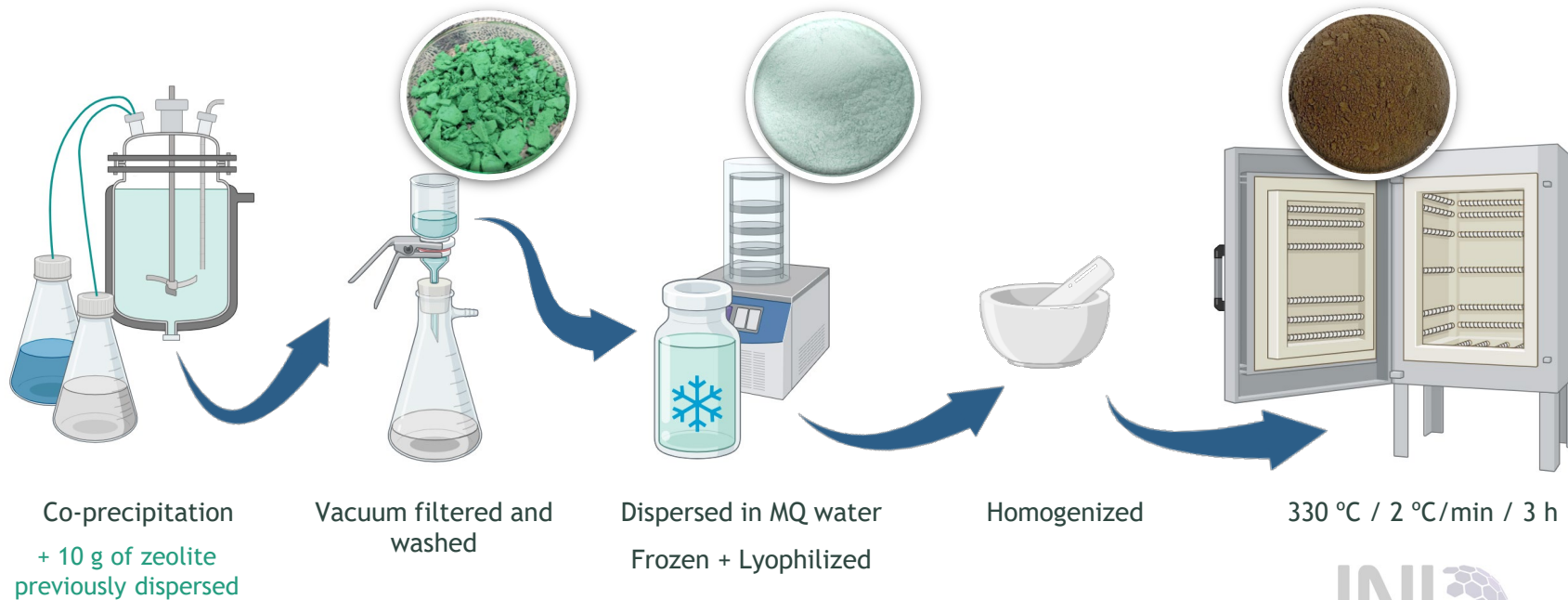
XRF data	Si (%)	Al (%)	O (%)	Si/Al ratio
SSZ-13	39.20	2.69	58.10	14.6

Crystallinity



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Tandem Catalyst Synthesis

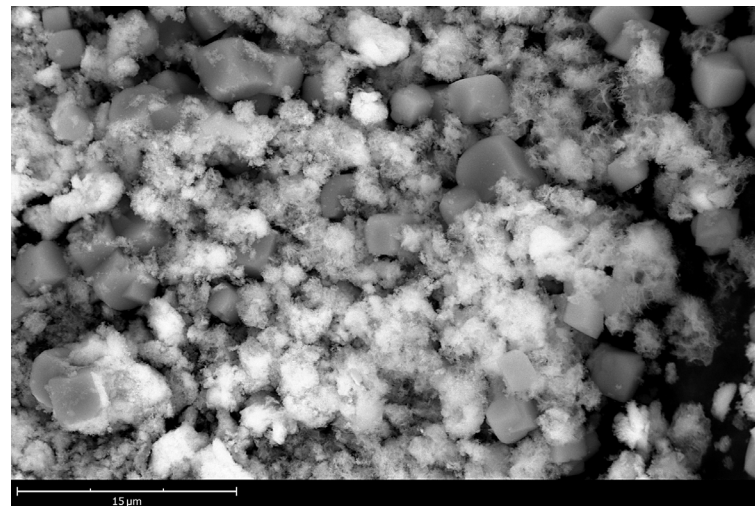
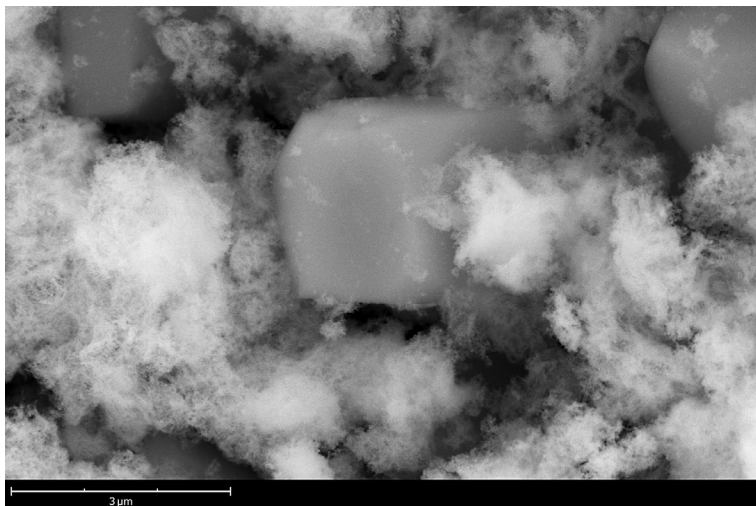


Tandem Catalyst Synthesis

Morphology and elemental composition

SSZ-13_CZA

Optimization strategy!



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XRF	Si (%)	Al (%)	Cu (%)	Zn (%)	O (%)	Oxide/zeolite
SSZ-13_CZA	21.80	2.12	27.20	11.20	37.70	61/39



Catalyst synthesis



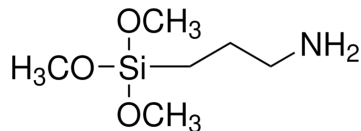
Tandem Catalyst Synthesis

Commercial SSZ-13 zeolite

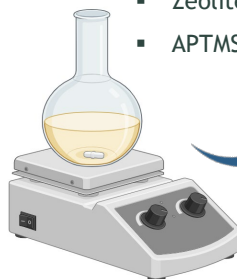
Functionalization as our optimization strategy

Can monoamine graft improve the CZA dispersion, and adhesion on the zeolite?

APTMS (3-aminopropyl)trimethoxysilane)



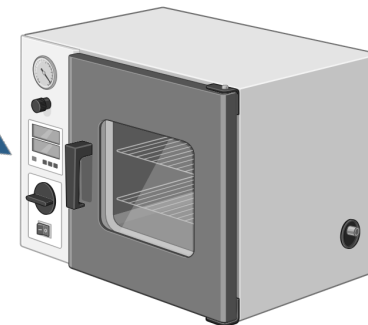
- Ethanol
- Zeolite
- APTMS



24 h - RT



Vacuum filtered
and washed

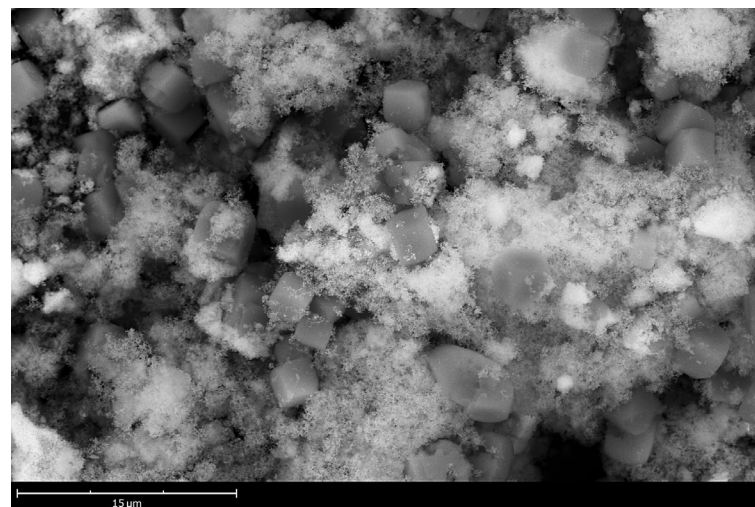
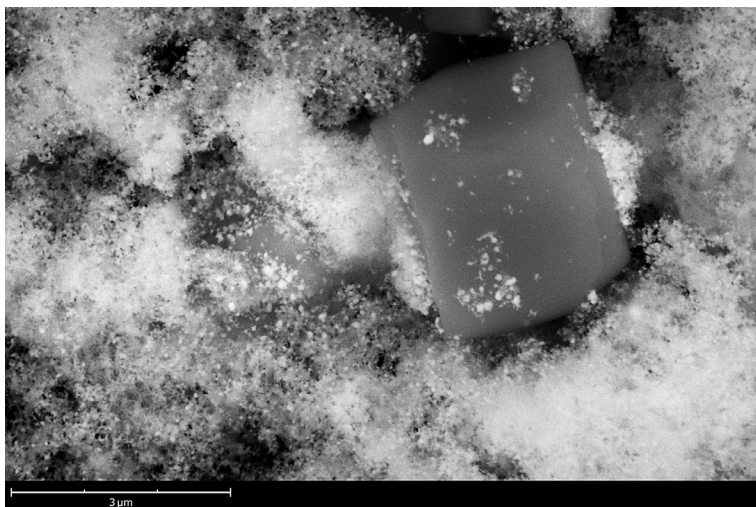


Overnight - 60 °C - vacuum

Tandem Catalyst Synthesis

Morphology and elemental composition **SSZ-13-F_CZA**

Optimization strategy!



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XRF	Si (%)	Al (%)	Cu (%)	Zn (%)	O (%)	Oxide/zeolite
SSZ-13_CZA-F	20.00	1.94	24.80	10.10	43.30	61/39

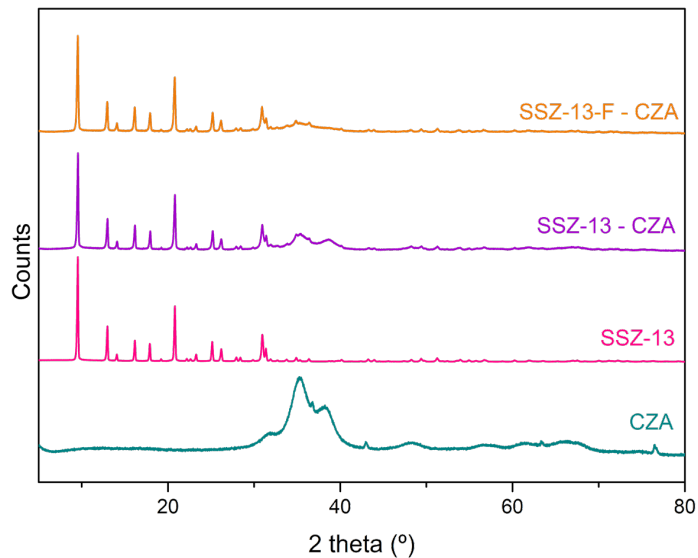


Catalyst characterization

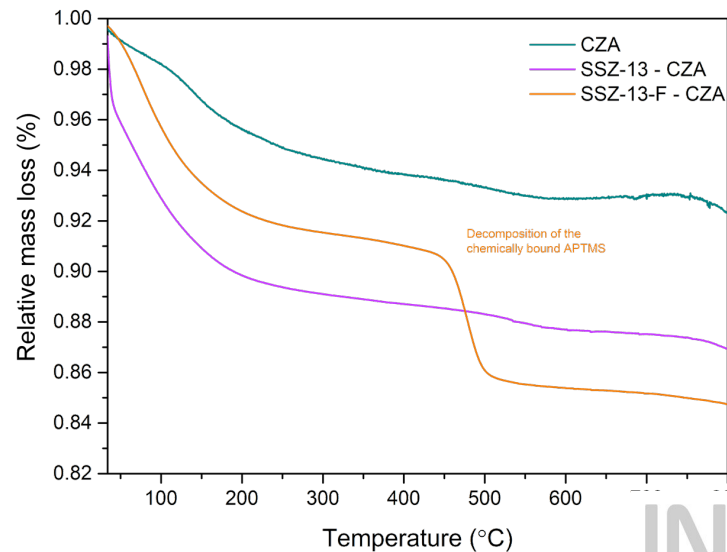


Tandem Catalyst Characterization

Crystalline structure



Thermostability





Results



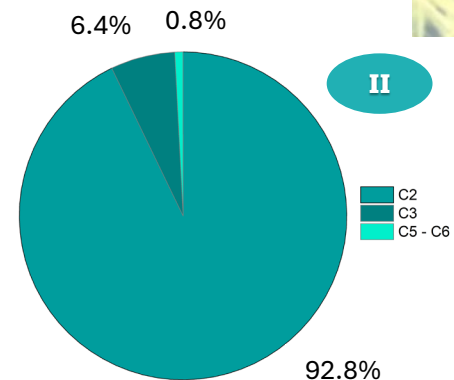
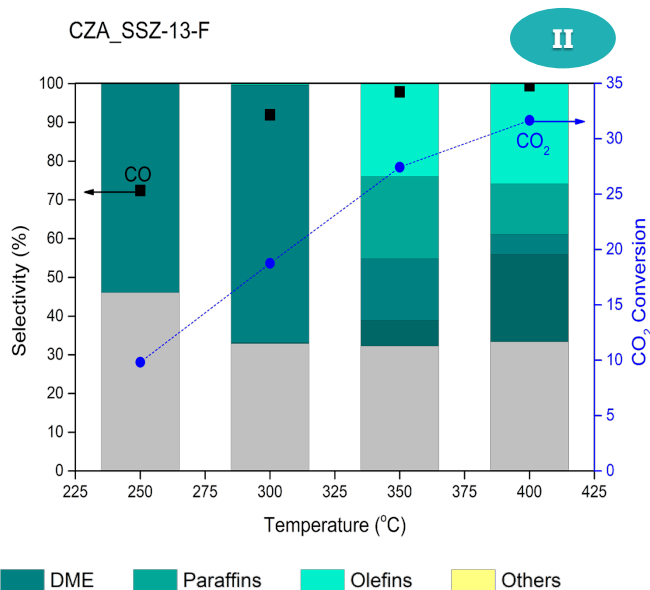
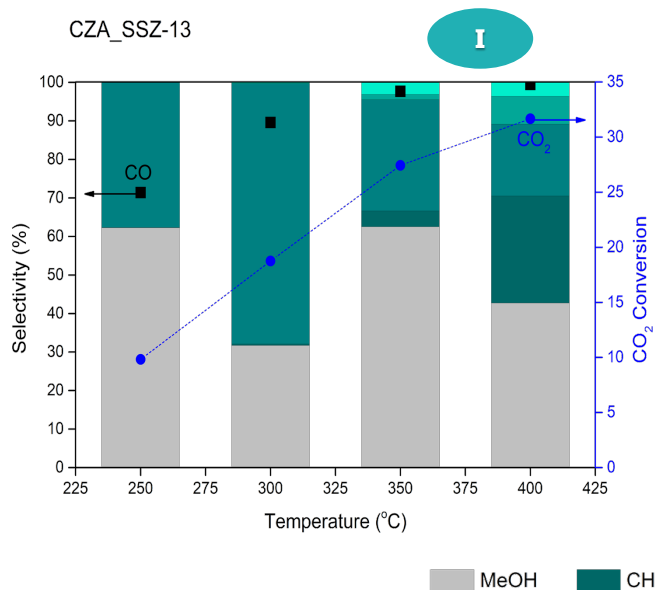
Tandem Activity

Catalytic tests



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MAX-PLANCK-GESELLSCHAFT

$S_{\text{olefins @ 350}^\circ\text{C}} = 23.9 \%$





Results



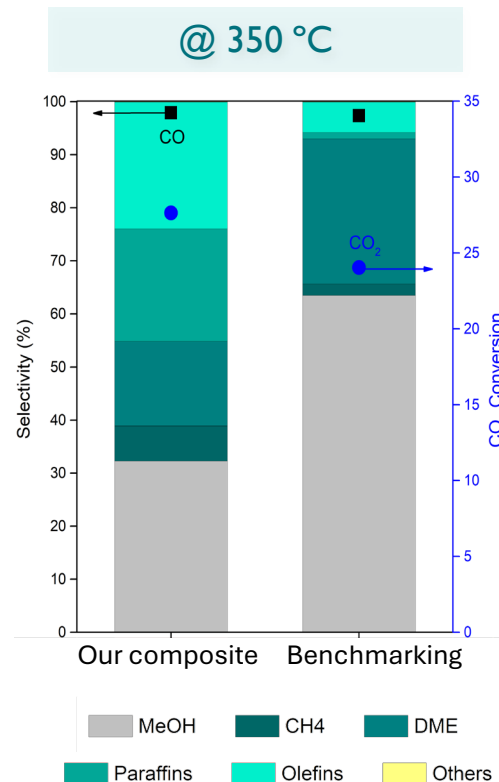
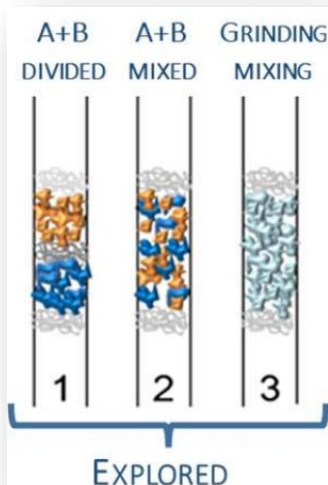
Tandem Activity

Catalytic tests



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Benchmarking
physical mixture





Catalyst synthesis

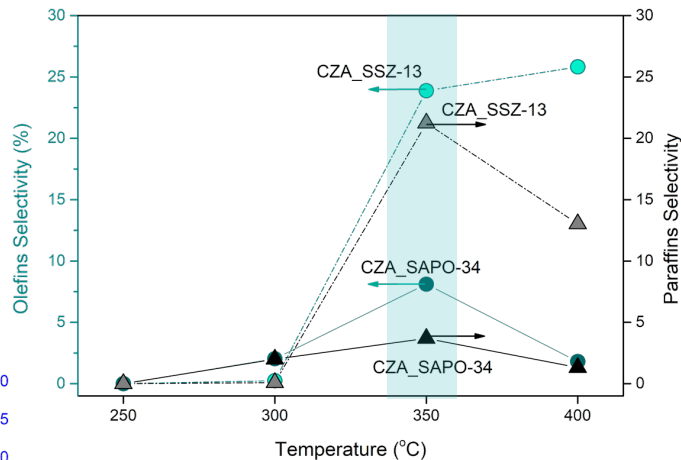
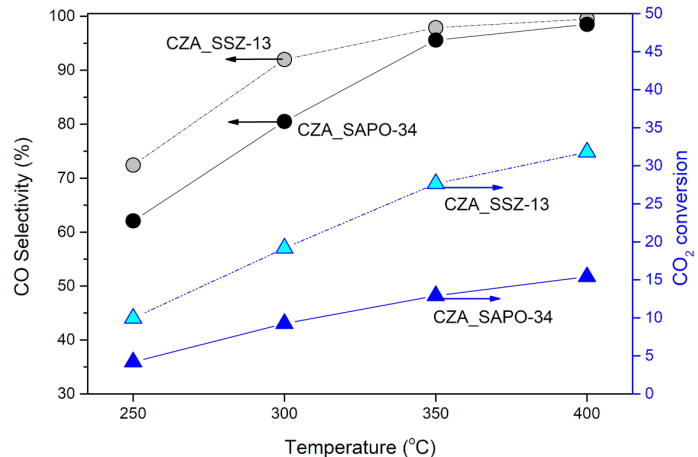


Tandem Catalyst

Materials selection: why not SAPO-34?

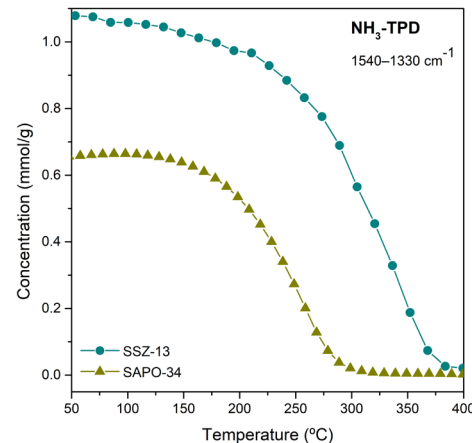
Composites:

- CZA_SSZ-13
- CZA_SAPO-34



Headroom on O/P

- Our SSZ-13 is low Si/Al
- Paraffins are tunable via Si/Al ratio
- SAPO-34 acidity is fixed



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Conclusions



MCs and MTO catalysts

Operating temperature range

Materials selection for best conversion and selectivity in the lowest temperature possible



Nanostructuring

Spatial distribution rule

Functionalization of the MTO catalyst, improved olefins production



Cat_A_{MeOH} / Cat_B_{MTO} ratio

Efficient matching rule

Equilibrium between MeOH formation and conversion, leading an increase in S_{olefins} at 350 °C!



The Future



Tune Si/Al ratio to 50 and 100 (MTO catalyst acidity control)



Improve spatial distribution by core-shell structuring

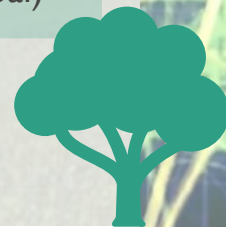


Incorporation into membrane reactor ($X \geq 80\%$ and $S \geq 95\%$ goal)



Towards green ethylene production...

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Thank you for your attention!

Contact: natalia.spera@inl.int



MEMCAT

<https://www.memcatproject.eu/>

KOLEN'KO GROUP 

<https://nanochemgroup.org/>

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MemCat Project

Experimental details



Catalytic activity

- Catalytic tests



- ✓ Activation: 250 °C (5 °C/min) for 2h with 10% H₂/Ar, total flow rate: 1460 NmL/min (~209 NmL/min per reactor)
- ✓ Reaction stream, CO₂ : H₂ → 1:3 (22.5 vol.% CO₂ / 67.5 vol.% H₂ / 10% Ar as internal standard)
- ✓ Analysis via GC/MS
- ✓ T = 150-400 °C, p = 30.0 bar
- ✓ Weight of catalyst: 0.5 g
- ✓ Total flow rate: 140 NmL/min per reactor - dwell: 12h in each temperature
- ✓ Weight Hourly Space Velocity (WHSV): 16800 ml g⁻¹ h⁻¹

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