Creating an Unconventional Type of Paper: Cellulose Microspheres and Pulp Fibers

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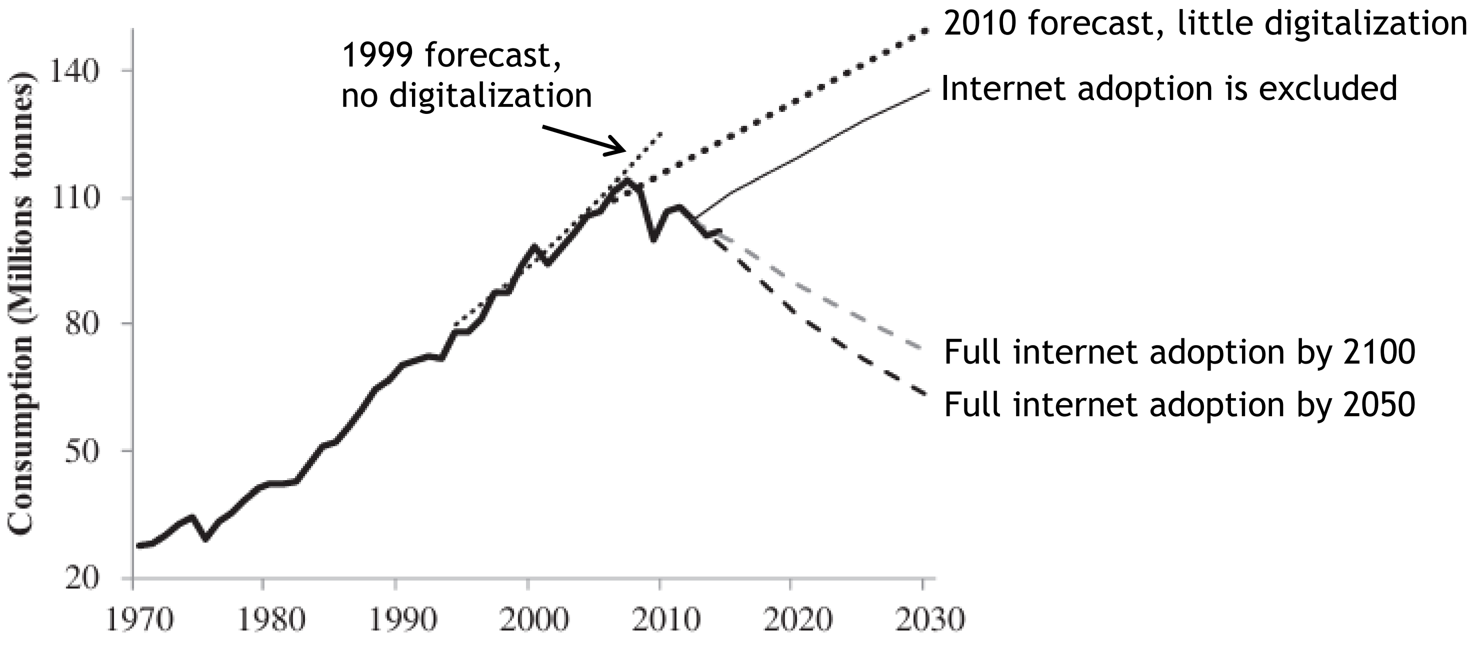
Why research in unconventional paper materials matters

Functional versatility: Paper can be engineered or modified to exhibit new functions

Sustainability and renewability: Paper is a biobased, renewable, and biodegradable material

Existing infrastructure: Well-established global production and recycling system for paper

Reduced demand for traditional paper products: Paper and paperboard consumption in Europe declined by 15.3% from 2022 to 2023



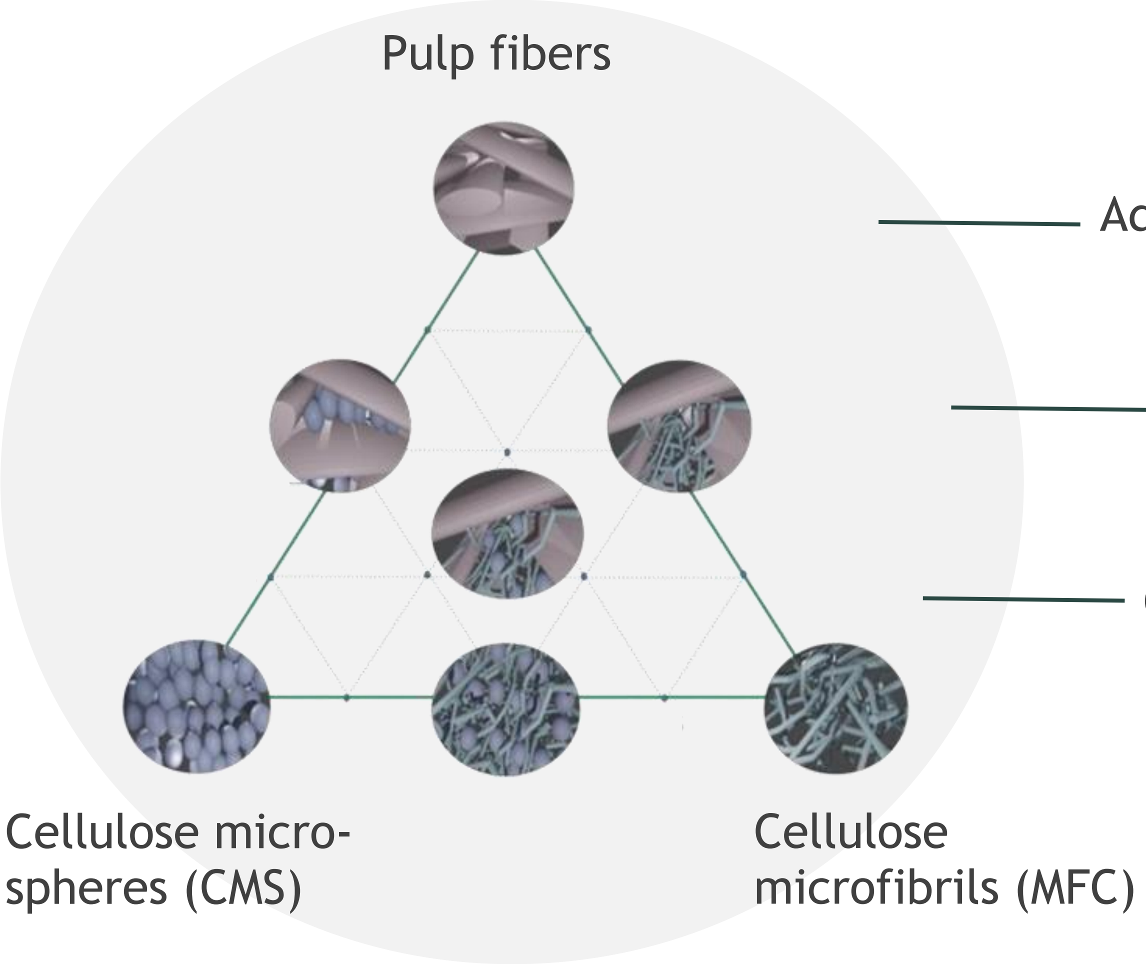
Emerging applications of paper

Strategies for paper development

- Incorporation of functional additives → Paper-based flexible electrode
- Conversion into functional materials → Pressure sensor for wearables
- Structure engineering → Paper separator for supercapacitors

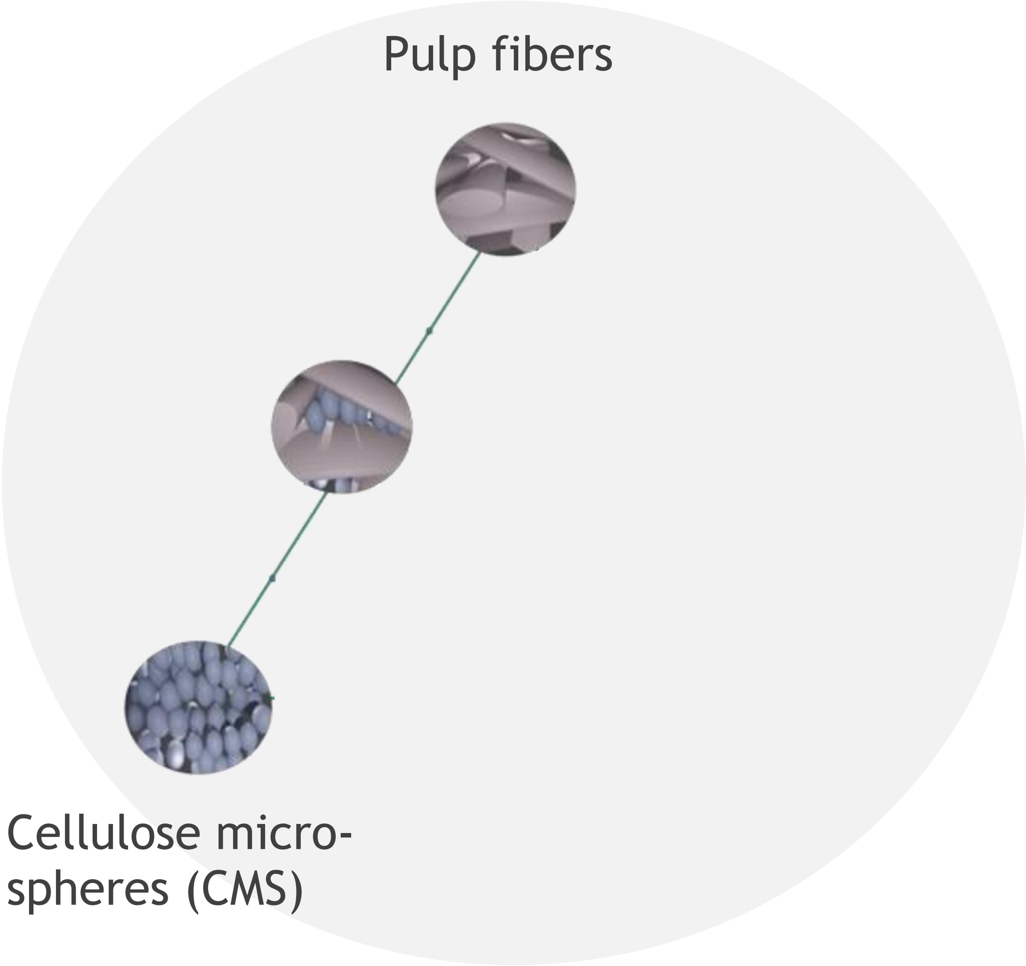
Liang, D. et al. (2023). Int. J. Biol. Macromol., 244, 125414 / Feng, X. et al. (2024). Mater. Chem. Phys., 321, 129489 / Selinger, J. et al. (2024). Carbohydr. Polym., 326, 121899

Structure engineering: Combining fibers with cellulose microspheres & MFC



- Adjustable properties
- Diverse microstructure
- Chemically identical

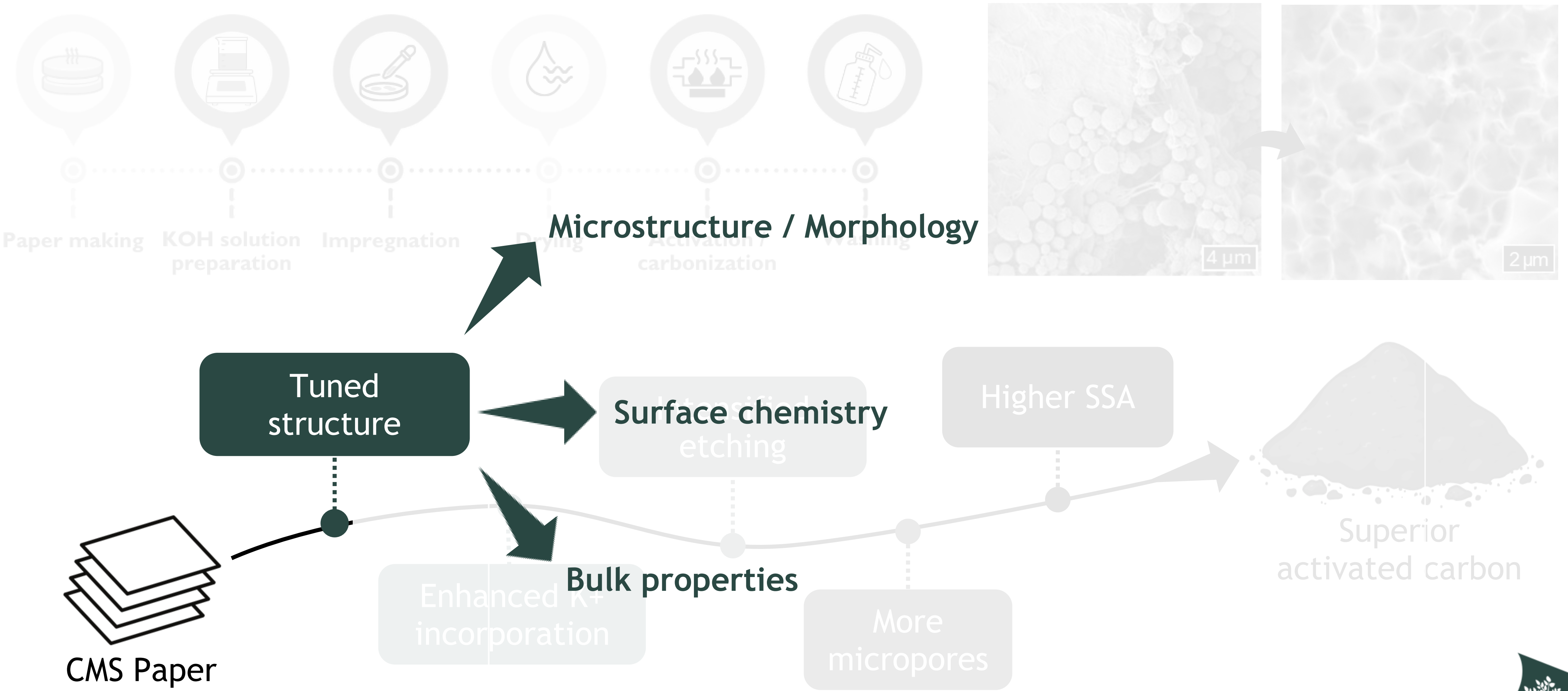
Structure engineering: CMS paper



Why integrate CMS into paper?

1. Microstructure tuning
2. Functionalization potential
3. **Utilizing the composite paper as a precursor for activated carbon synthesis**

From CMS paper to activated carbon with thermal KOH activation

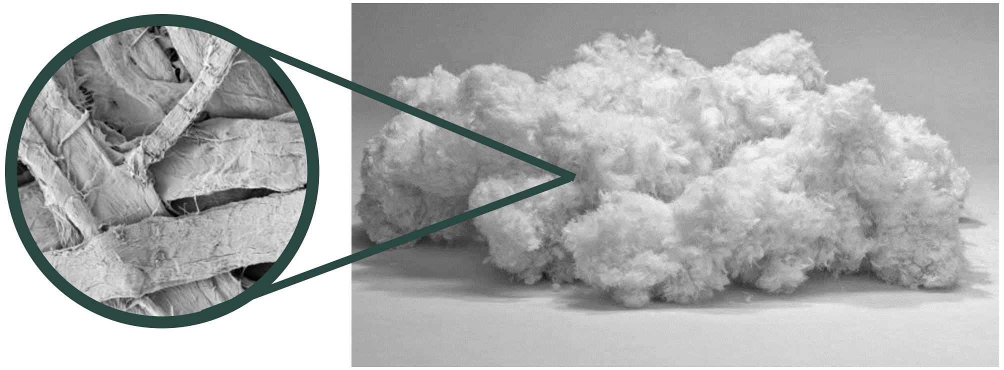


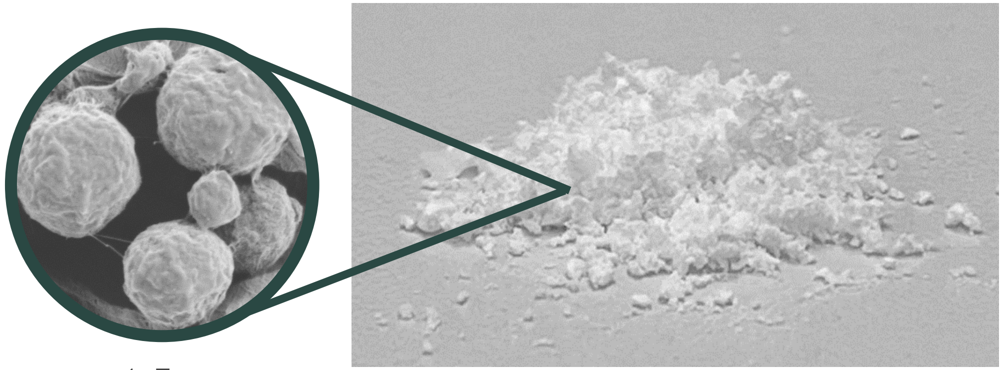
Raw materials

+ Chemically pure (dissolving-grade)
- Poor paper strength

l =722 µm
w = 20 µm

Bleached hardwood pulp
(acid sulfite)





⌀ 1.5 µm

Cellulose microspheres
(CMS)

Regenerated
cellulose

?

Scheer, A. et al. (2024). Carbohydr. Polym., 338, 122207

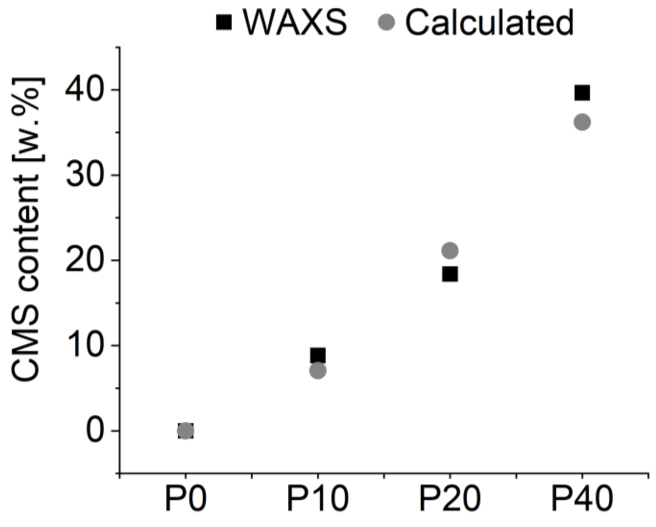
Hand sheet production

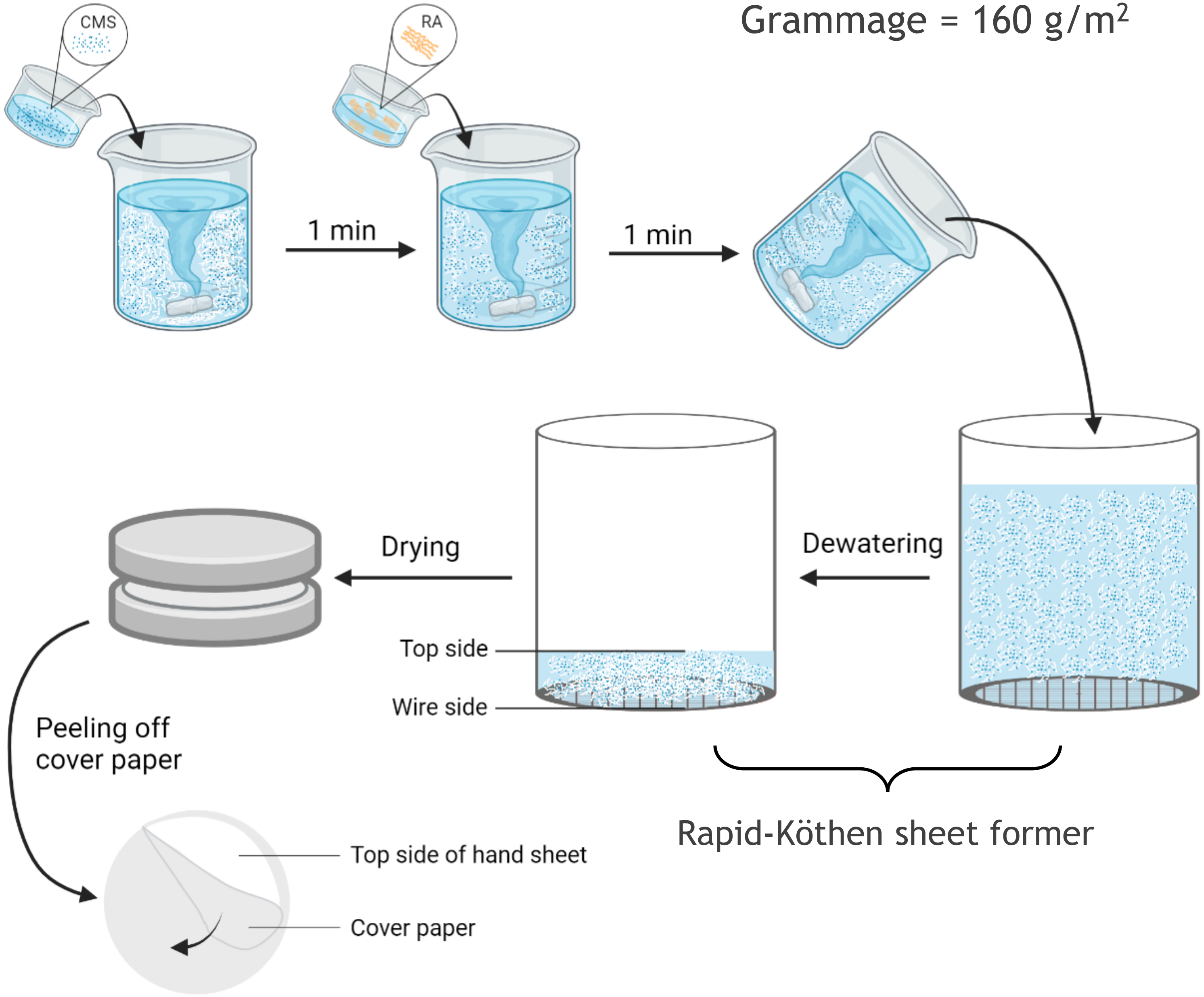
Grammage = 160 g/m^2

$$actual\ CMS\ mass\ in\ the\ sheet = sheet\ mass\ - fiber\ mass$$

$$CMS\ retention = \frac{\boldsymbol{actual\ CMS\ mass}}{added\ CMS\ mass}$$

$$CMS\ content = \frac{\boldsymbol{actual\ CMS\ mass}}{sheet\ mass}$$





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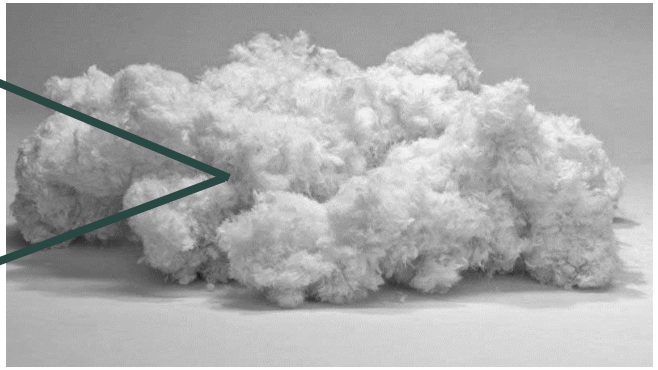

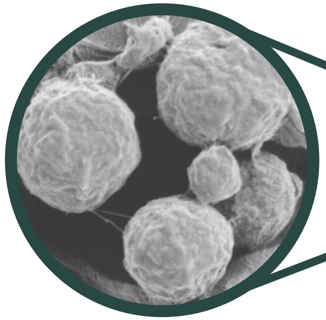

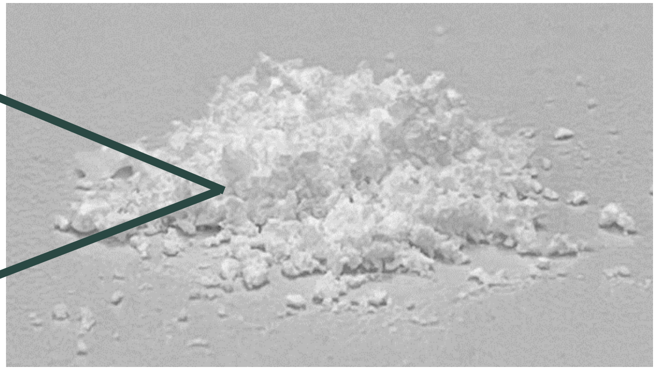

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Cellulose microspheres
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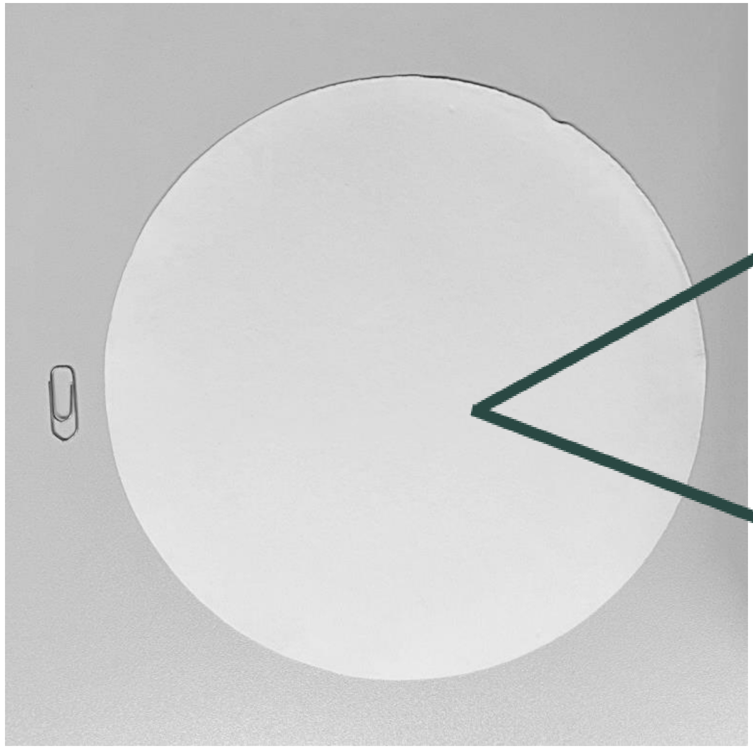
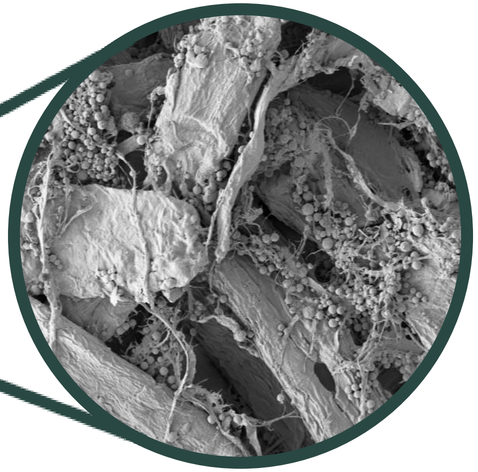
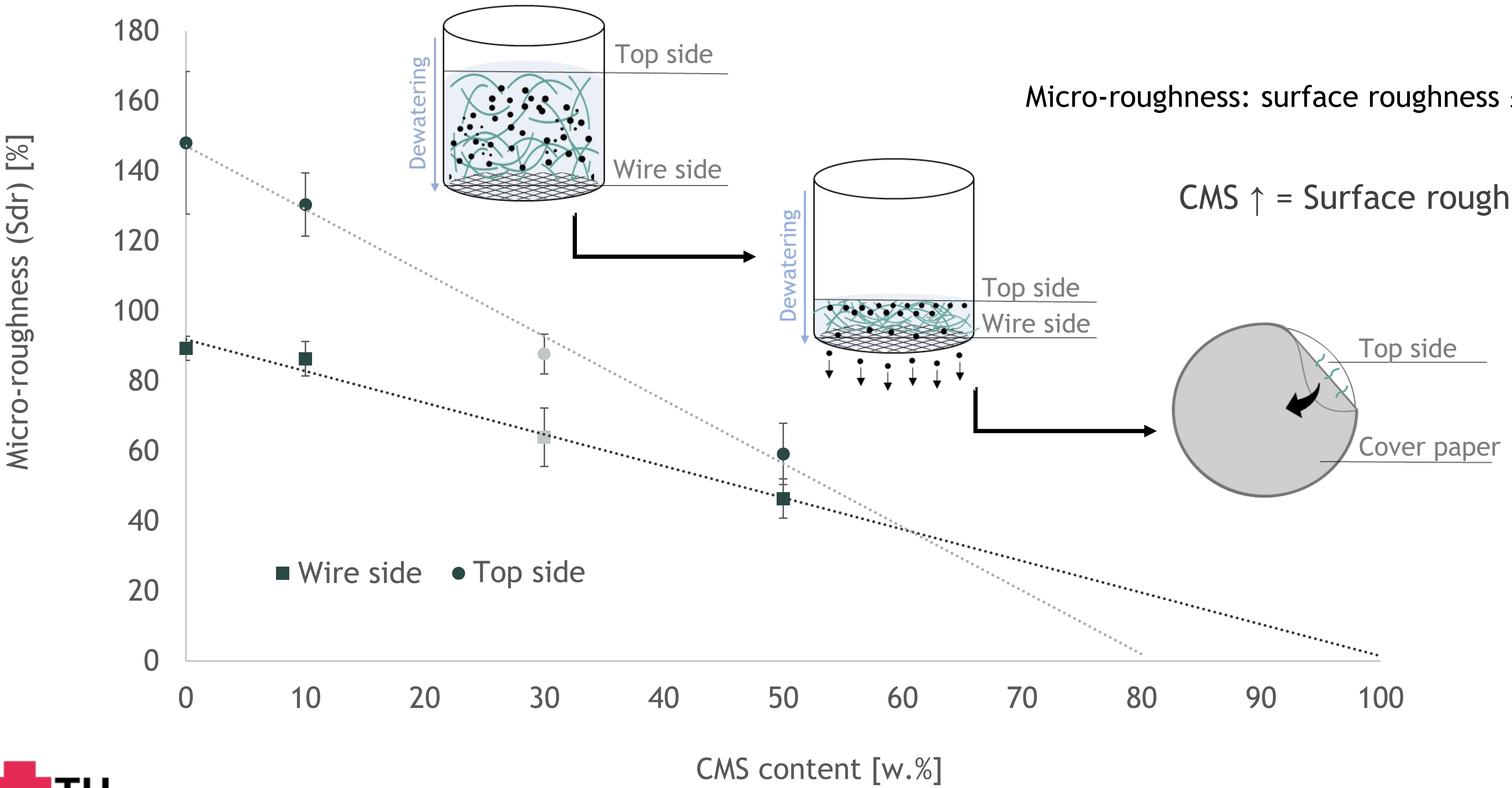




CMS Paper

Surface properties

Surface roughness by infinite focus microscopy



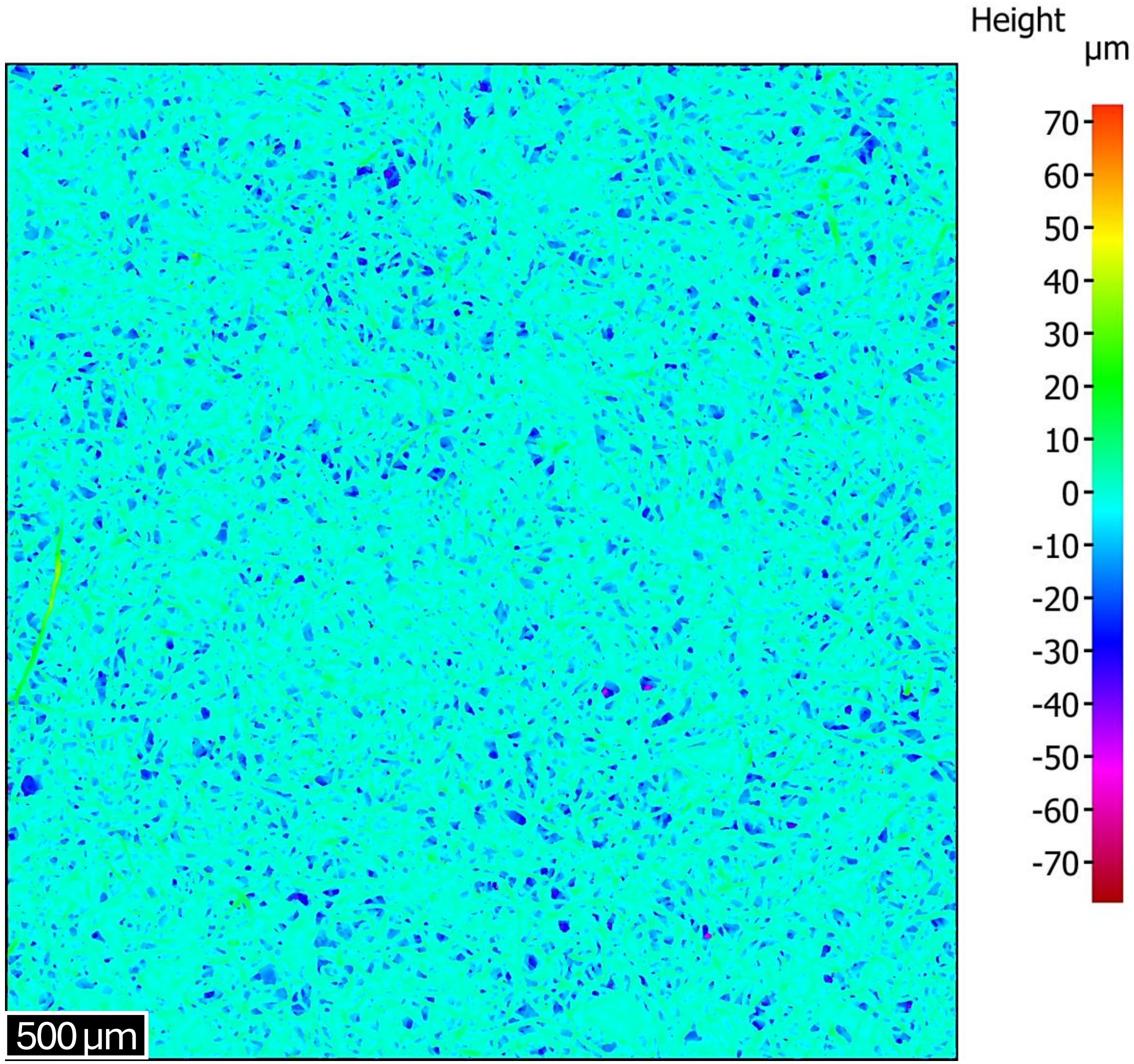
Micro-roughness: surface roughness ≤ 100 µm

CMS ↑ = Surface roughness ↓

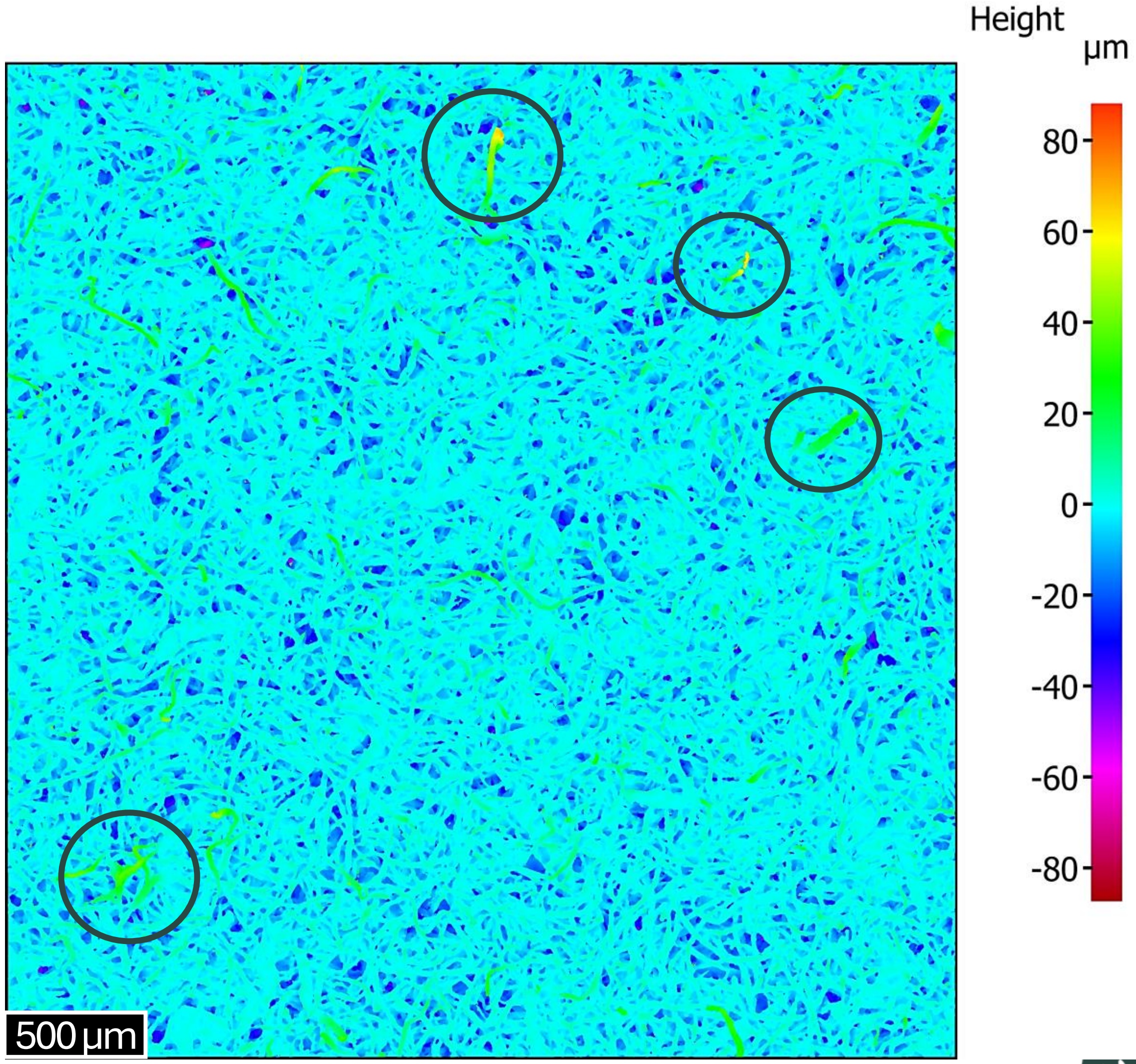
Scheer, A. et al. (2024). Carbohydr. Polym., 338, 122207

Sdr - value = Percentage deviation from a surface with a micro-roughness of 0 µm

Surface roughness of fiber-only paper

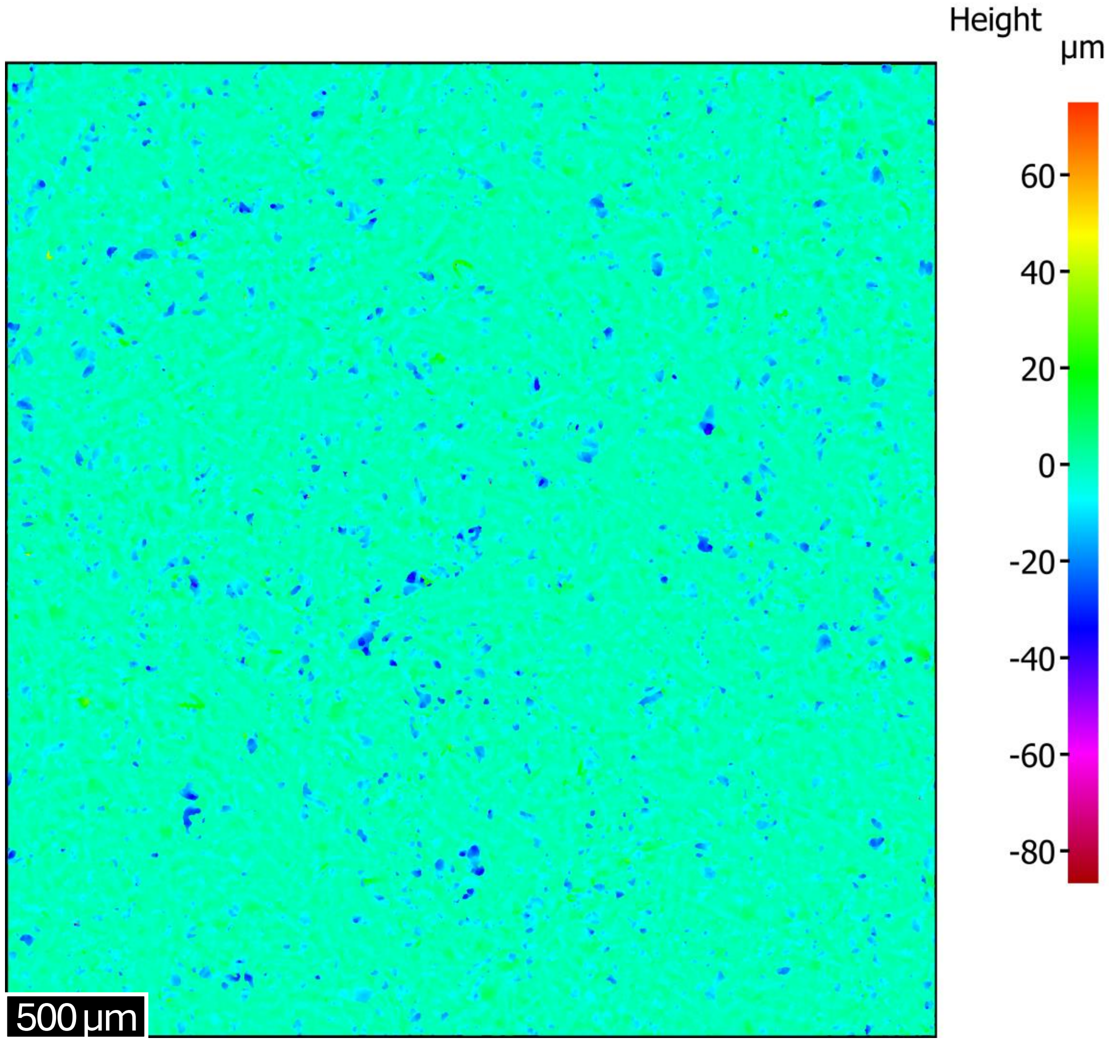


Wire side

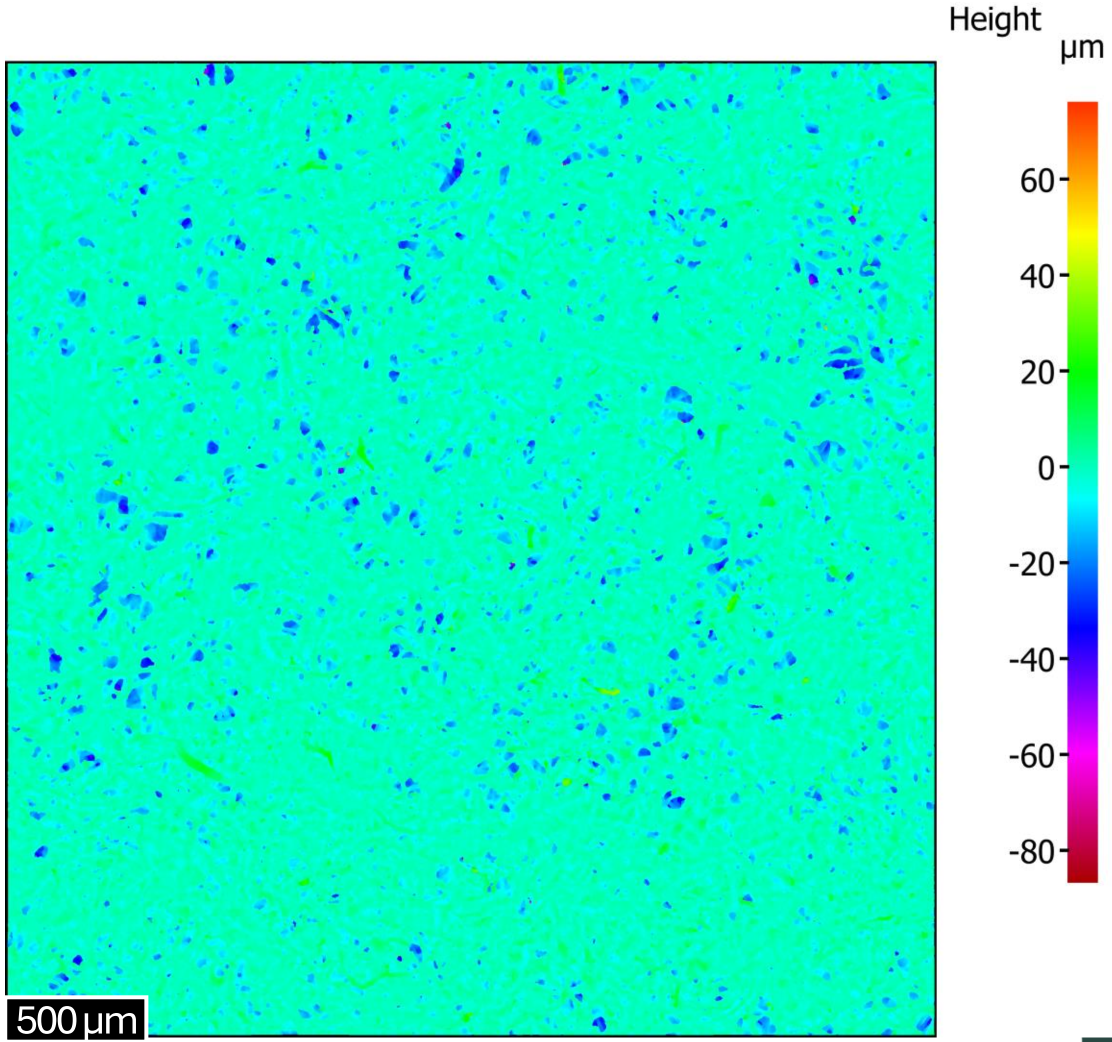


Top side

Surface roughness of 50 w.% CMS paper



Wire side



Top side

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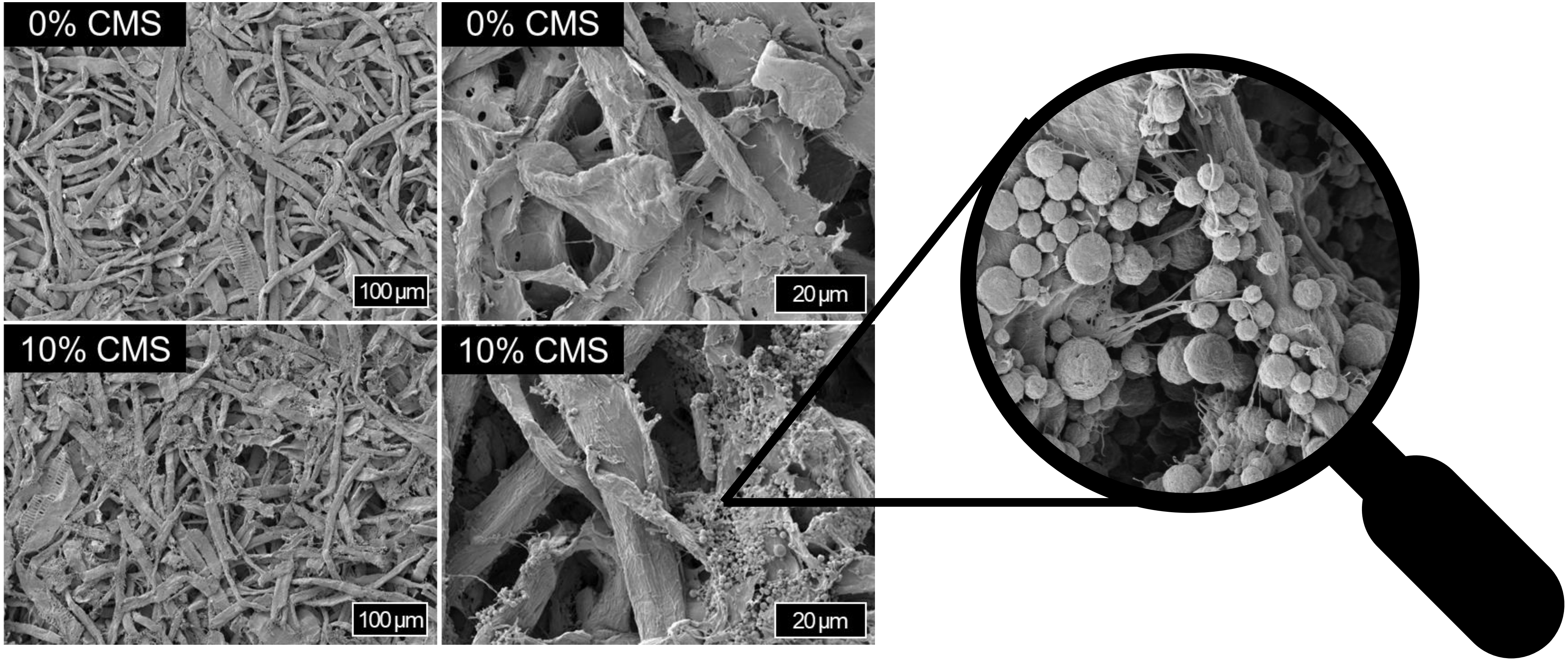
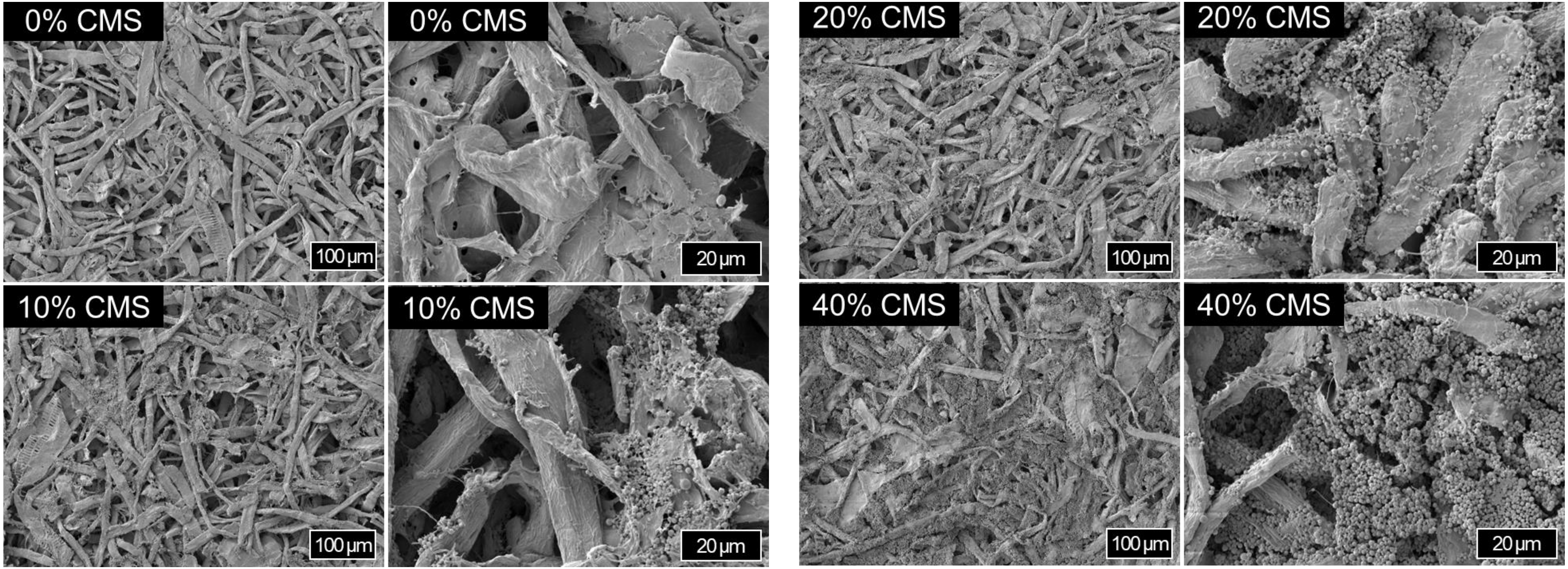
Surface microstructure



CMS adhere to fibrils, partially cover fiber surfaces, form agglomerates, and fill interfiber voids

Surface microstructure



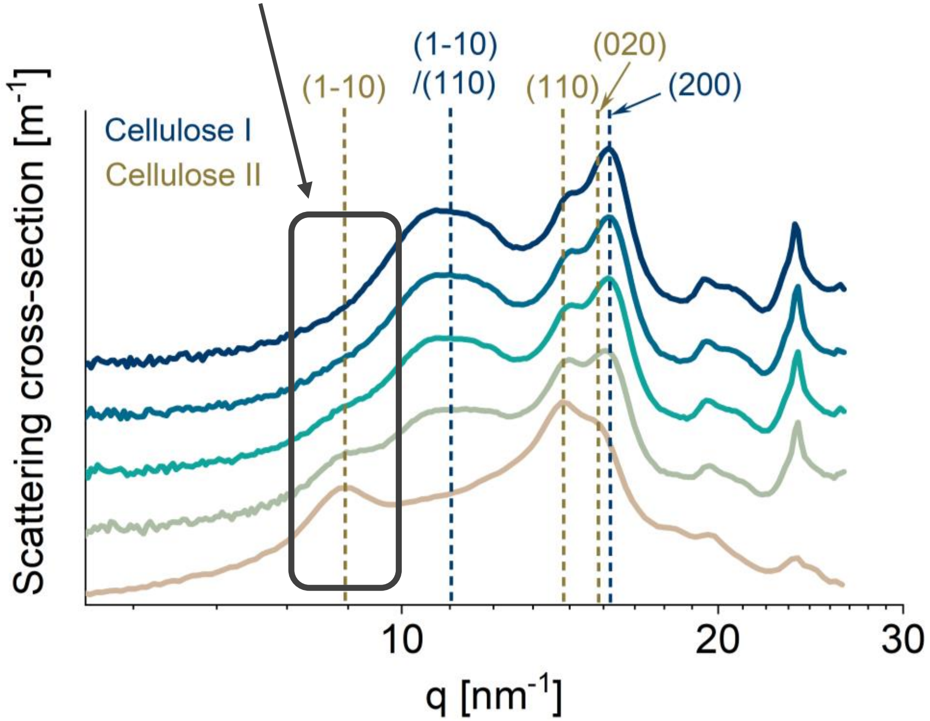
CMS adhere to fibrils, partially cover fiber surfaces, form agglomerates, and fill interfiber voids

Scheer, A. et al. (2025). Carbon, 121036.

Surface chemistry / crystallinity by WAXS & FTIR

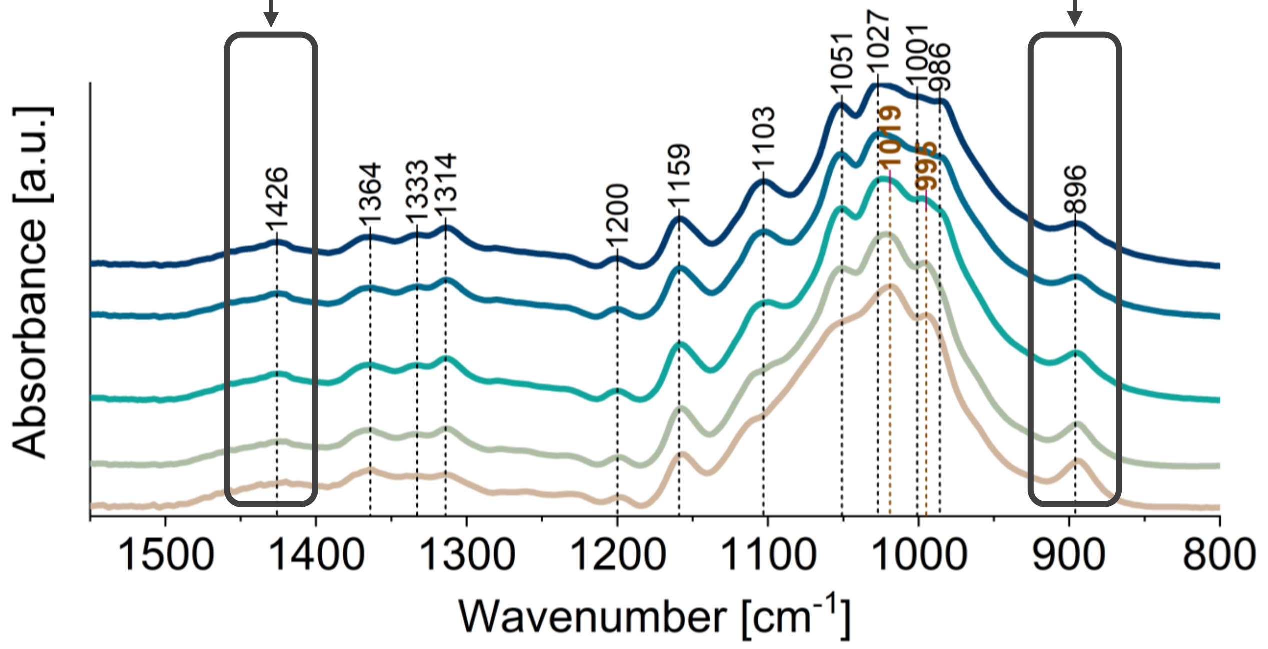
0 w.% CMS   10 w.% CMS   20 w.% CMS   40 w.% CMS   100 w.% CMS

Antiparallel chain packing
& expanded intersheet spacing



Crystallinity band,
CH_2 Scissoring

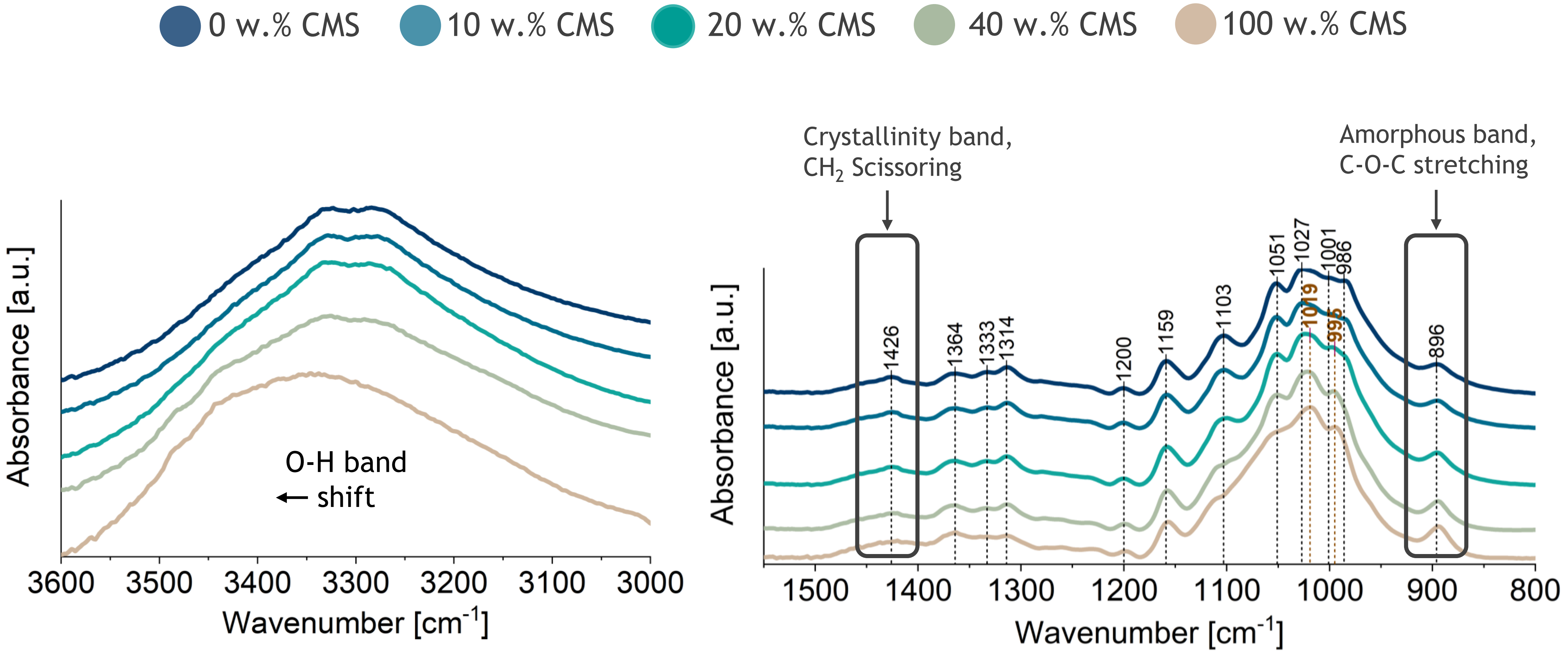
Amorphous band,
C-O-C stretching



CMS content ↑ = Amorphous character ↑

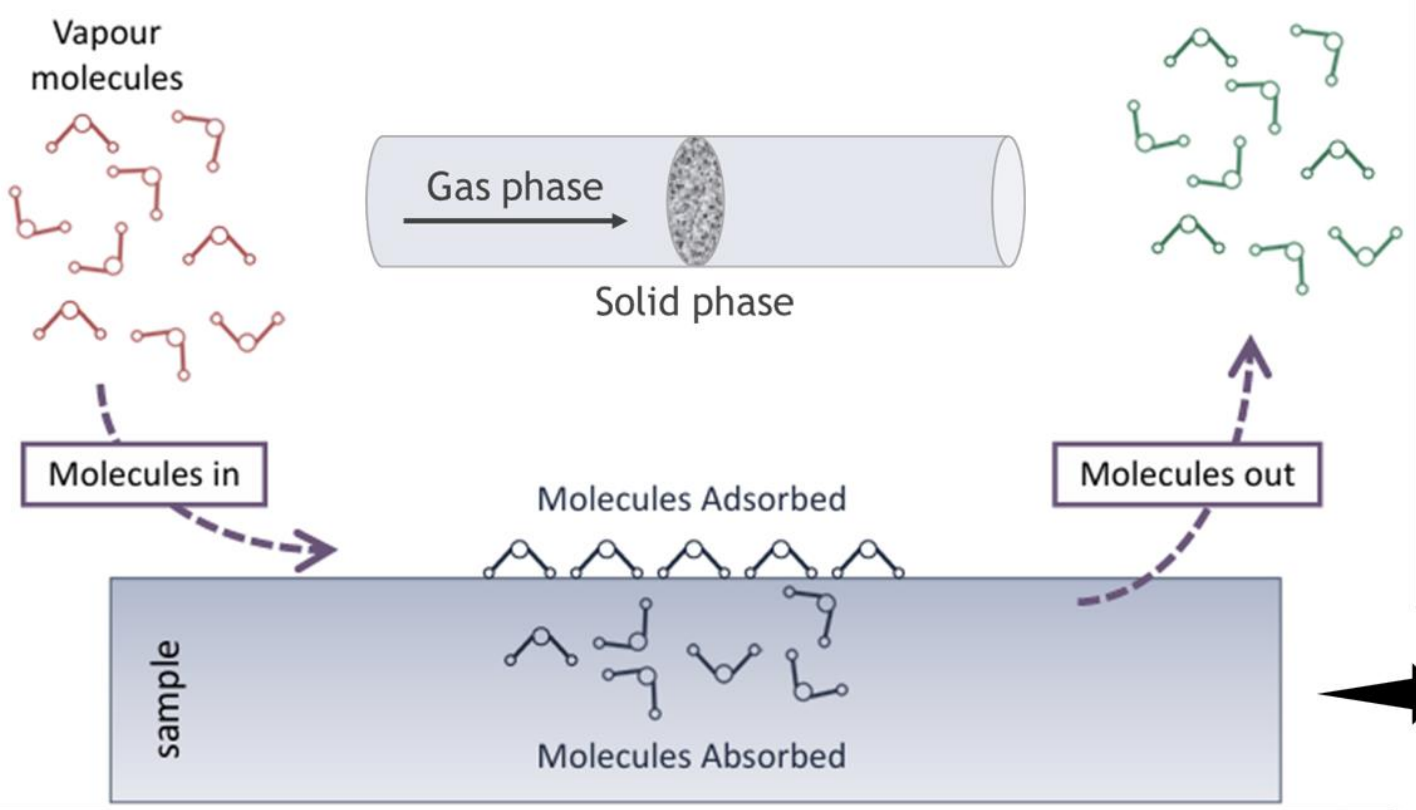
Scheer, A. et al. (2025). Carbon, 121036.

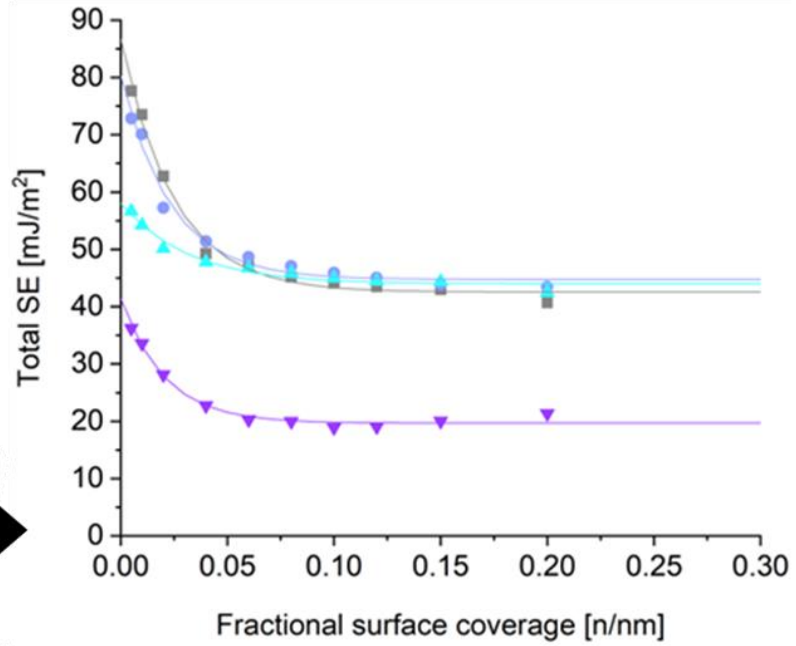
Surface chemistry & crystallinity by WAXS & FTIR



CMS content ↑ = Amorphous character ↑ O-H band shift ↑

Surface chemistry by Inverse Gas Chromatography (IGC)

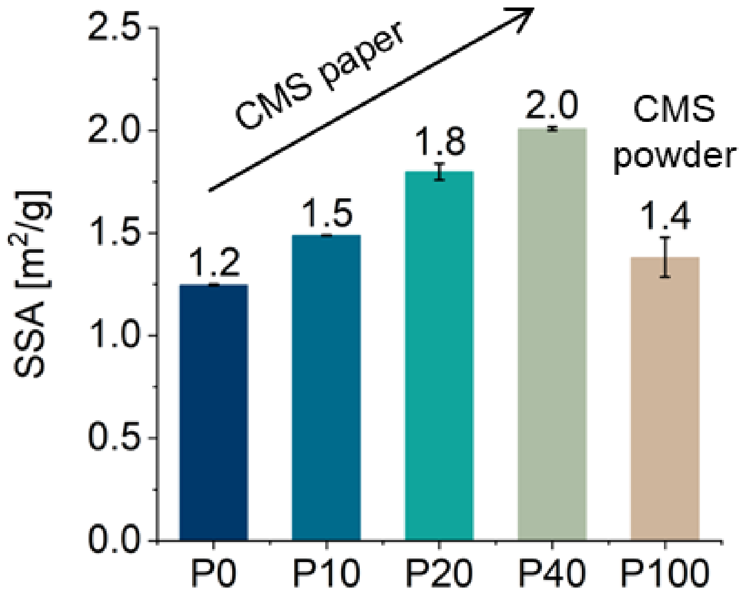


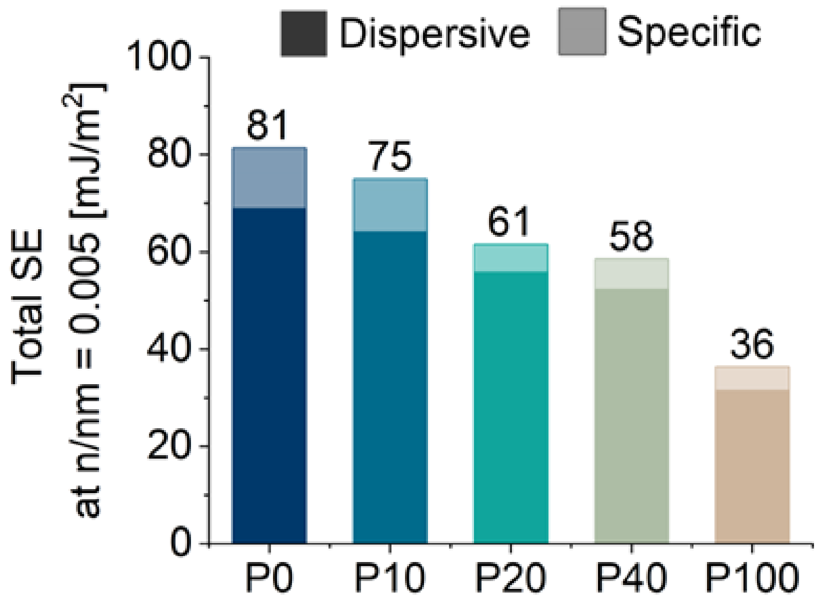


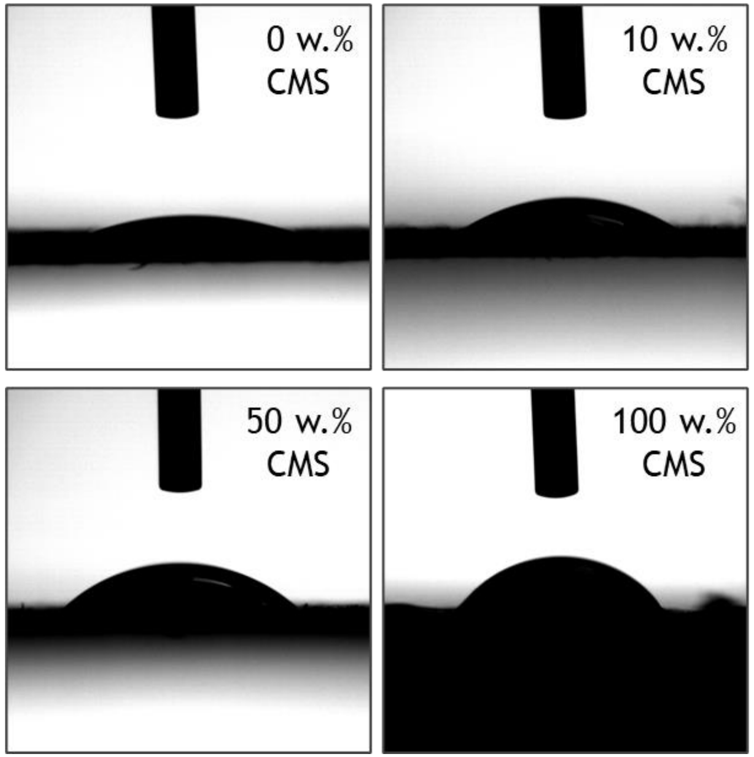
Series of n-alkanes for dispersive component, polar probes for specific component

Surface chemistry by Inverse Gas Chromatography (IGC)

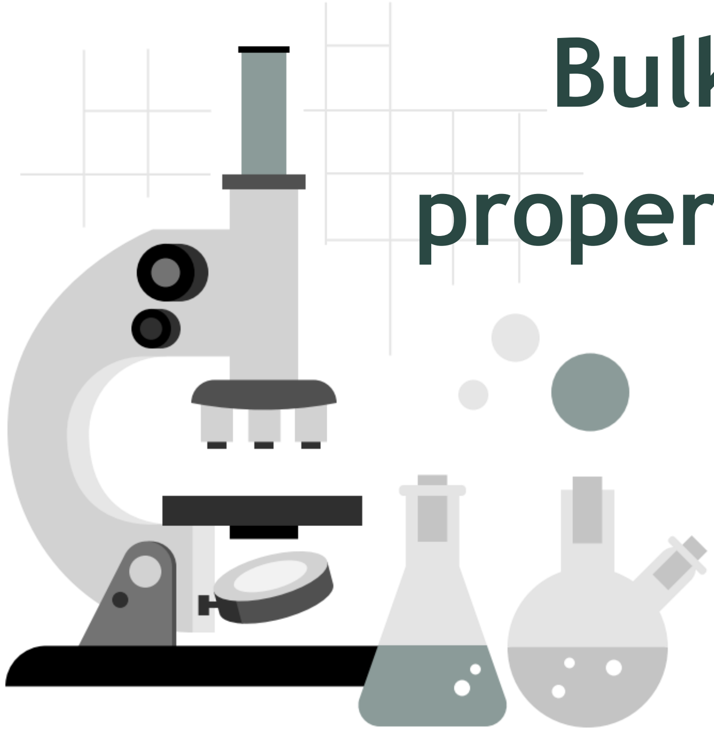
0 w.% CMS 10 w.% CMS 20 w.% CMS 40 w.% CMS 100 w.% CMS





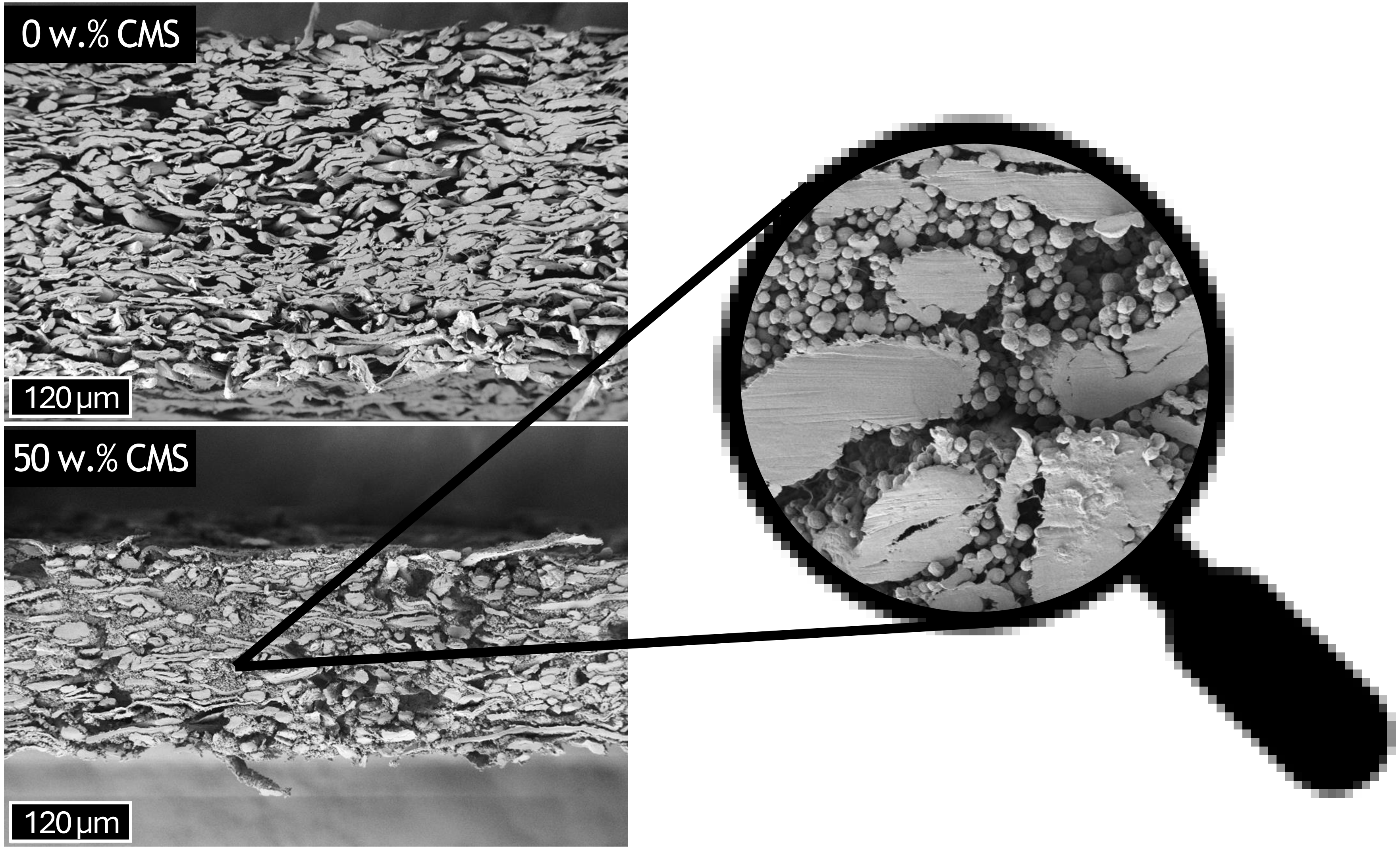


CMS ↑ = SSA ↑ Surface energy ↓ Wettability with water ↓



Bulk properties

Paper cross-section



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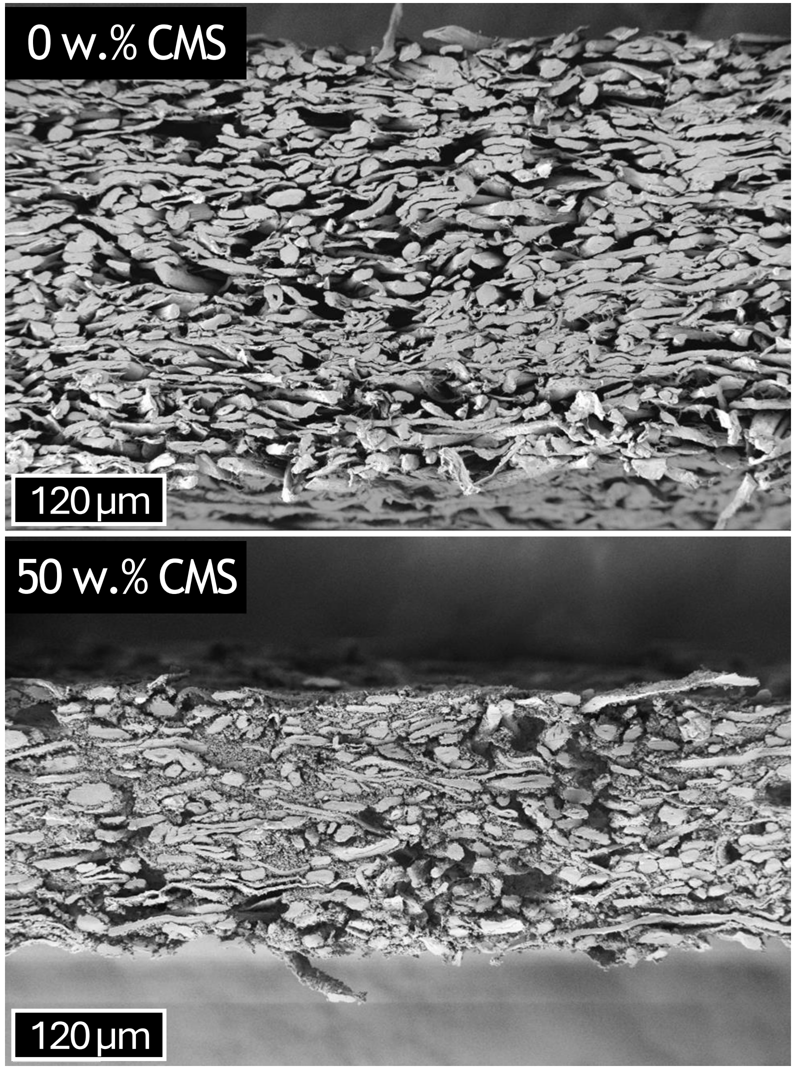
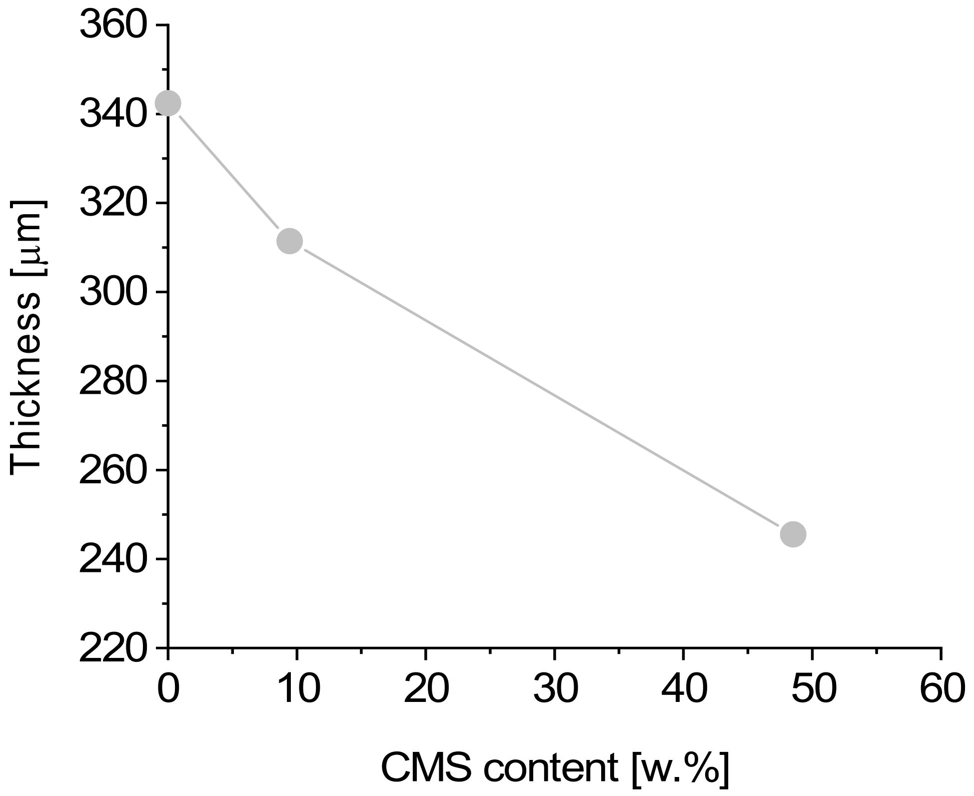
Paper cross-section





CMS in paper ↑ = Thickness ↓

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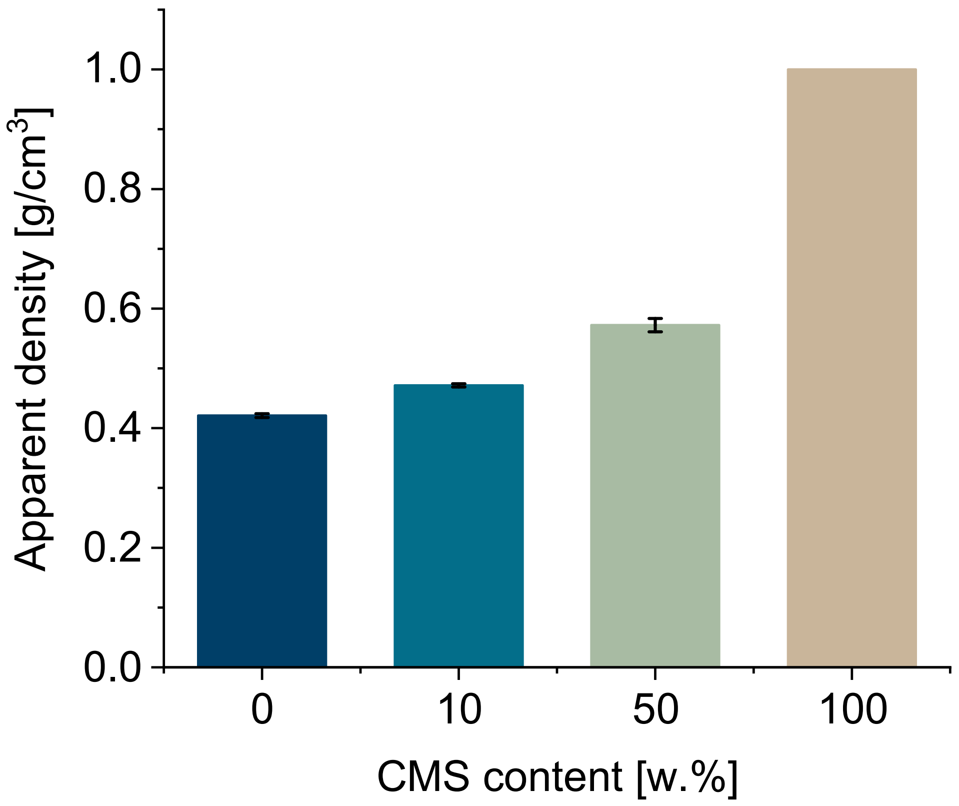
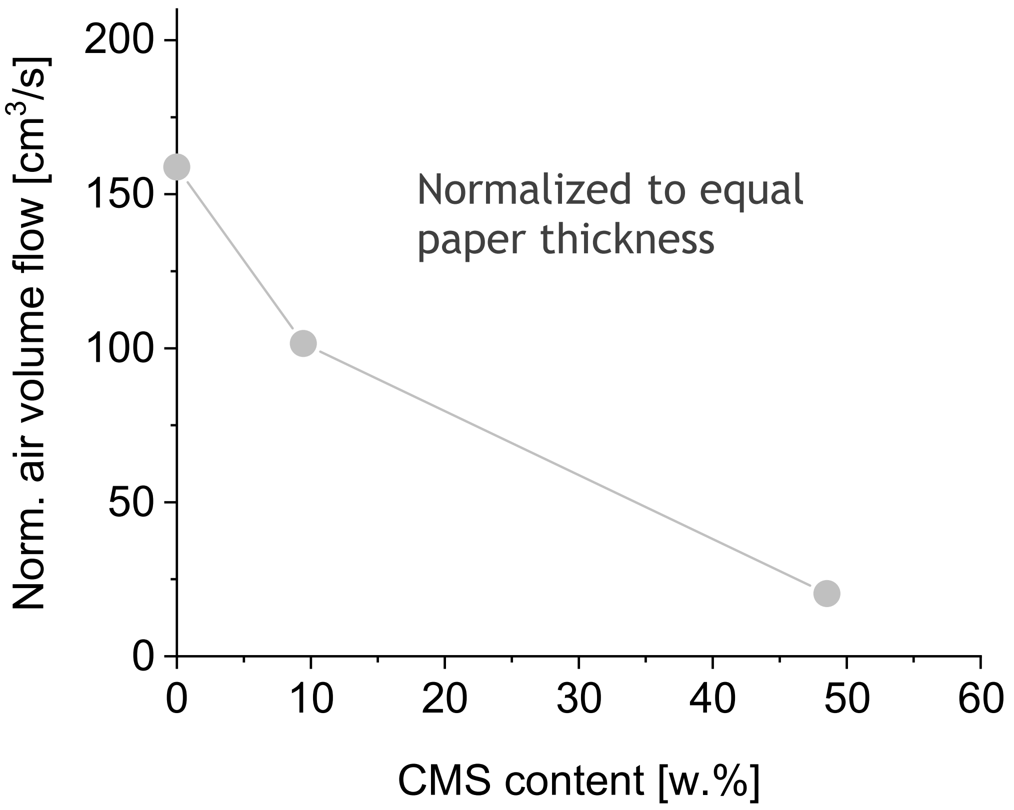
Paper density



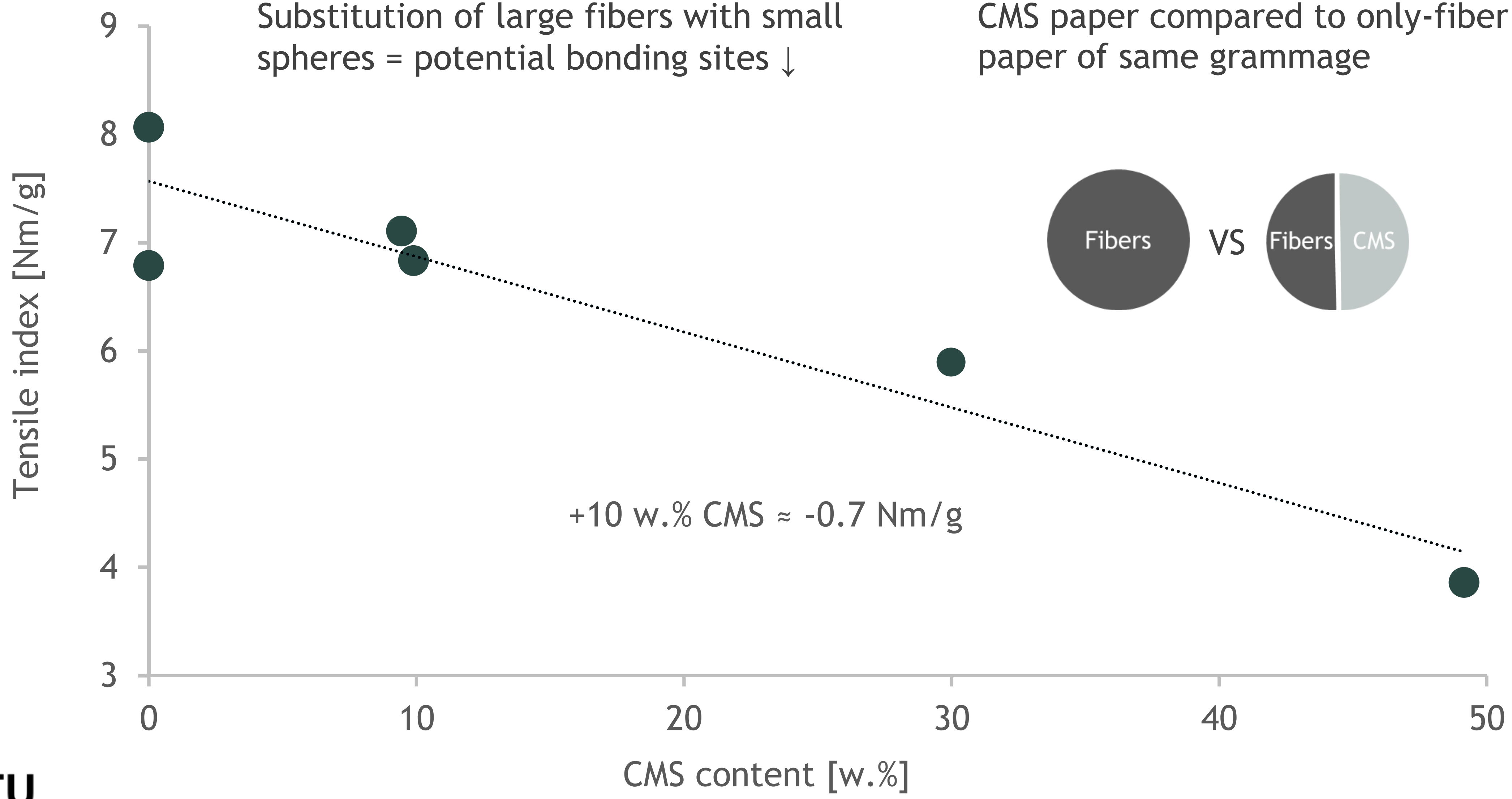


Closure of interfiber voids with CMS increases density and air tightness

Paper strength

Substitution of large fibers with small spheres = potential bonding sites ↓

CMS paper compared to only-fiber paper of same grammage



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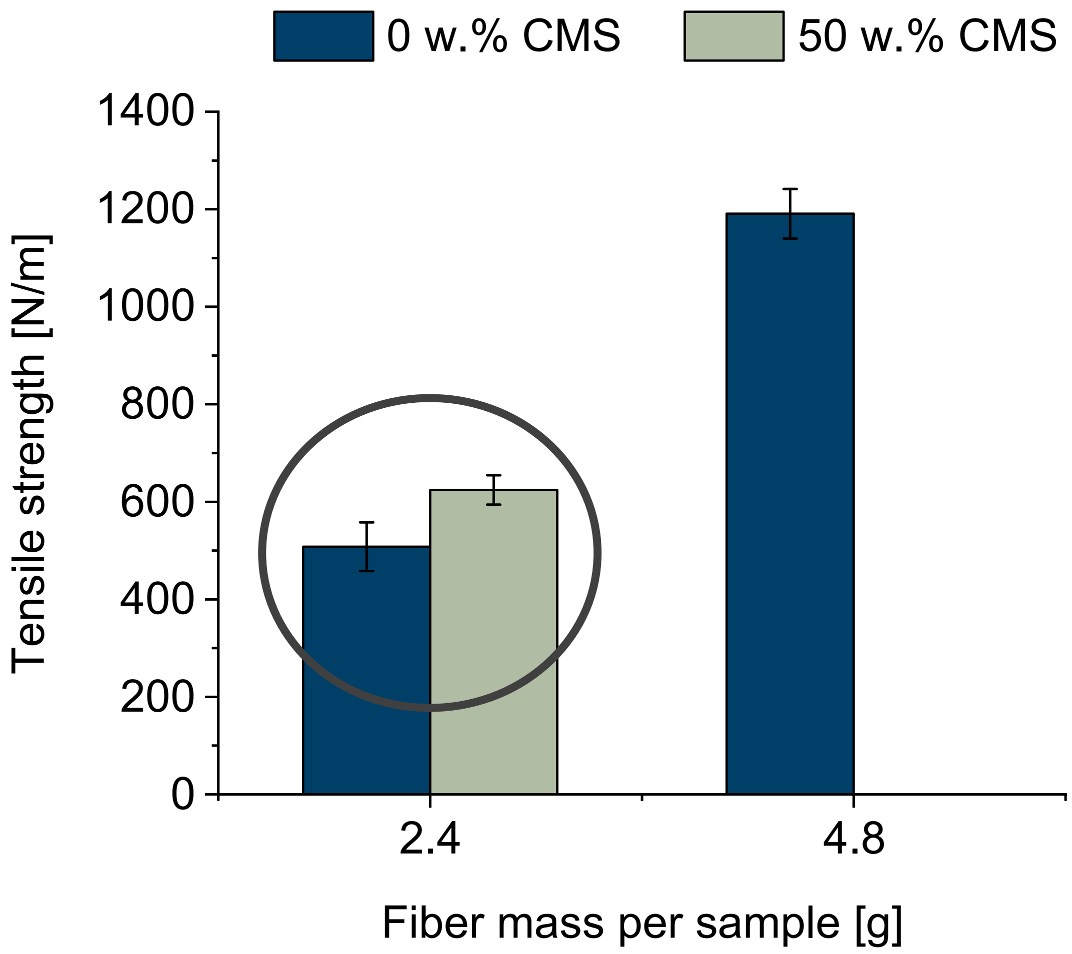
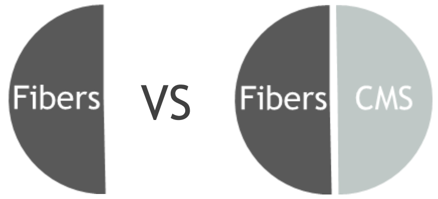
Paper strength



CMS paper compared to only-fiber paper of same fiber mass



Tensile strength:

CMS paper > fiber-only paper of same fiber mass

CMS addition does not negatively affect fiber-fiber bonding

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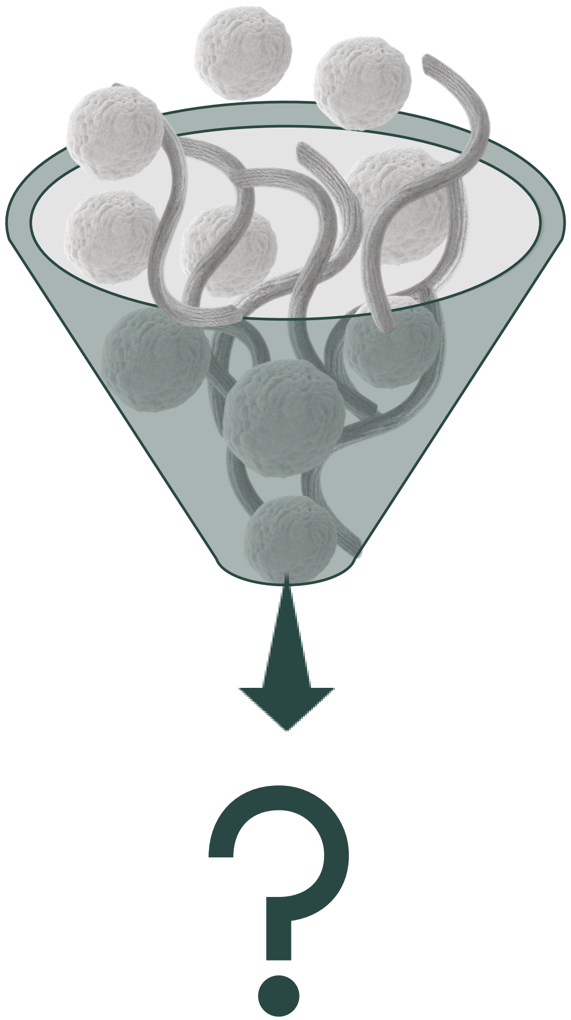
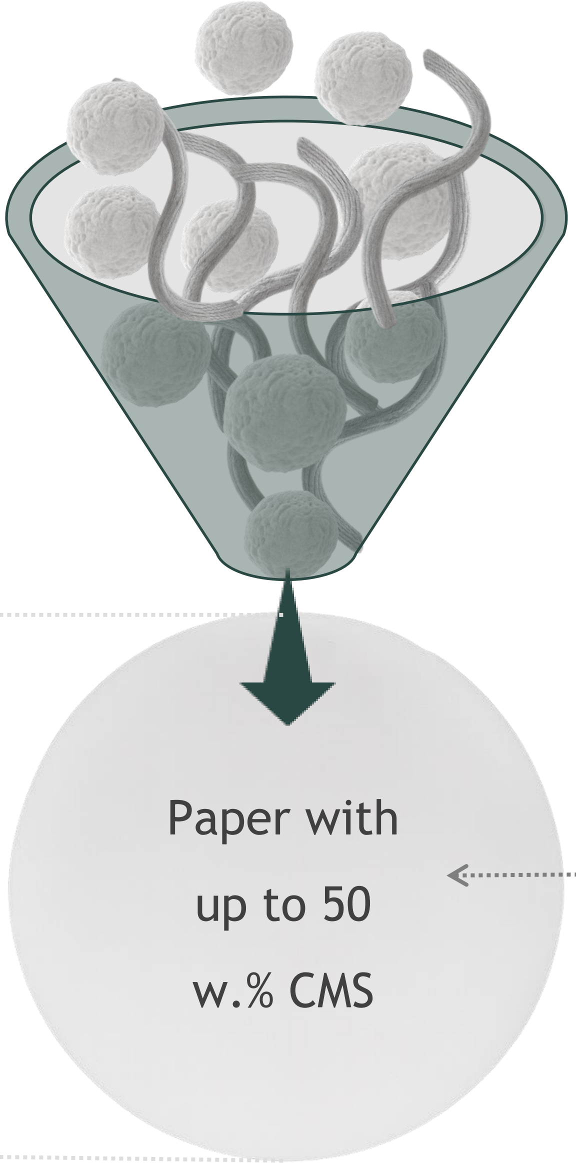
Conclusion



Conclusion

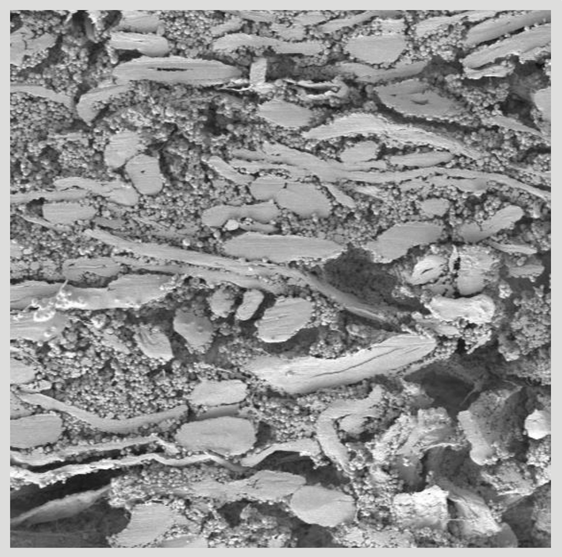


Thickness ↓

Density ↑

Air permeability ↓

Tensile strength ~



Microstructural complexity ↑

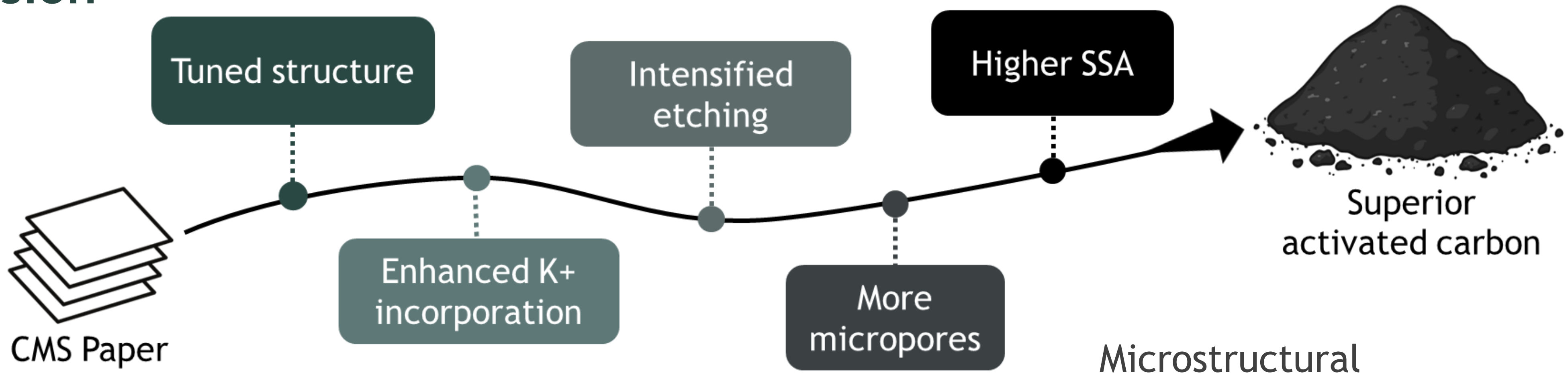
Surface roughness ↓

Crystallinity ↓

Surface area ↑

Surface energy ↓

Conclusion



Microstructural complexity ↑

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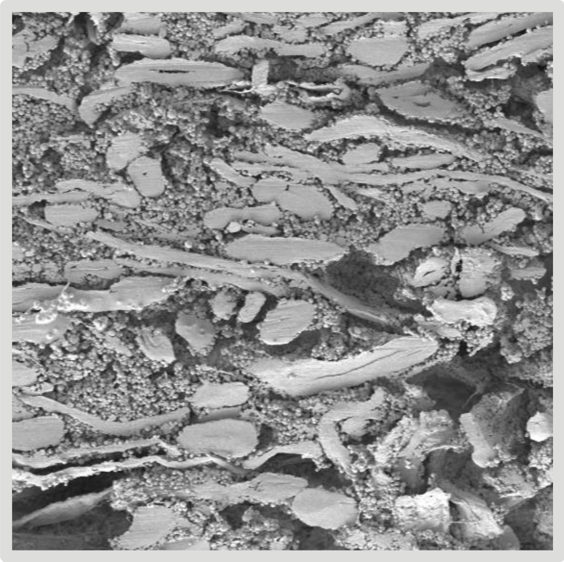
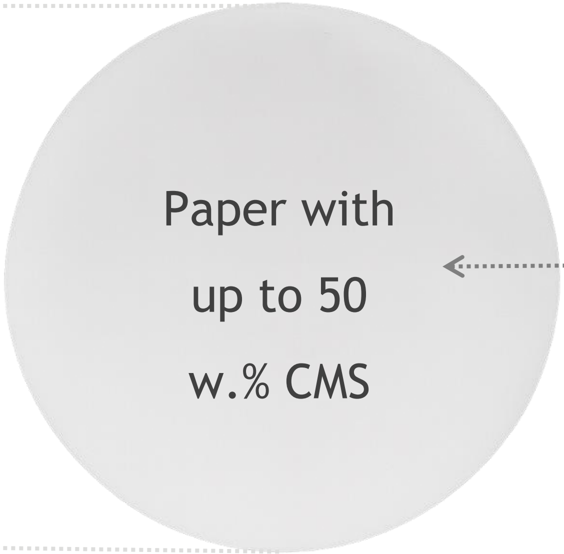
Tensile strength ~





Surface roughness ↓

Crystallinity ↓

Surface area ↑

Surface energy ↓

Conclusion

Ways in which CMS could add new functions to paper:

- Serving as carriers for functional materials
- Providing additional adhesion / interaction sites

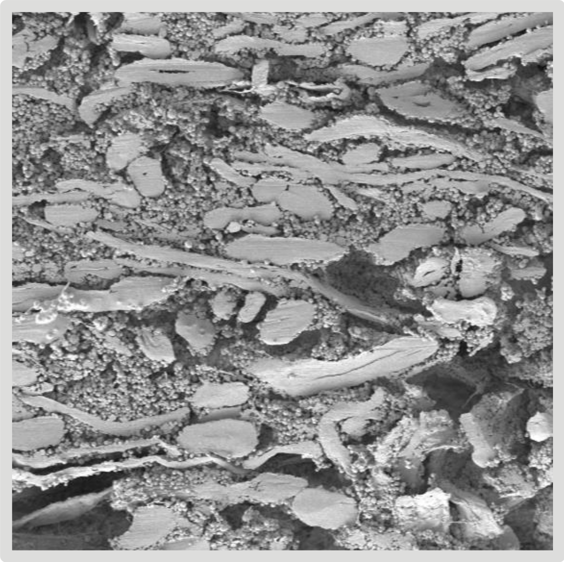
Microstructural complexity ↑

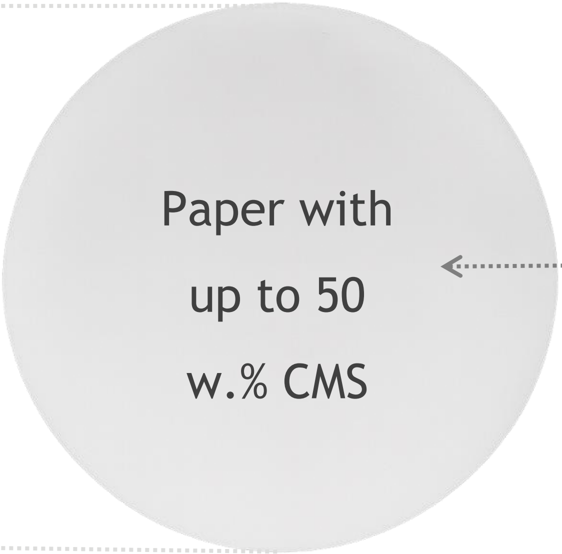
Thickness ↓

Density ↑

Air permeability ↓

Tensile strength ~





Surface roughness ↓

Crystallinity ↓

Surface area ↑

Surface energy ↓

Special thanks to …

- Stefan Spirk, Julian Selinger, lab technicians & my working group (TU Graz)
- Wolfgang Bauer (TU Graz)
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- FWF & DFG for funding “Cellstor“

TECHNISCHE UNIVERSITÄT DRESDEN

DFG Deutsche Forschungsgemeinschaft German Research Foundation

FWF Österreichischer Wissenschaftsfonds

Thank you for your attention!