Towards Correlative X-Ray Tomographic Imaging of Membranes for Improving Food Processing

G. Rudolph, J. Thuvander, E. Larsson, S. Hall, T. Nilsson Pingel, Pablo Villanueva-Perez, F. Lipnizki

Outline

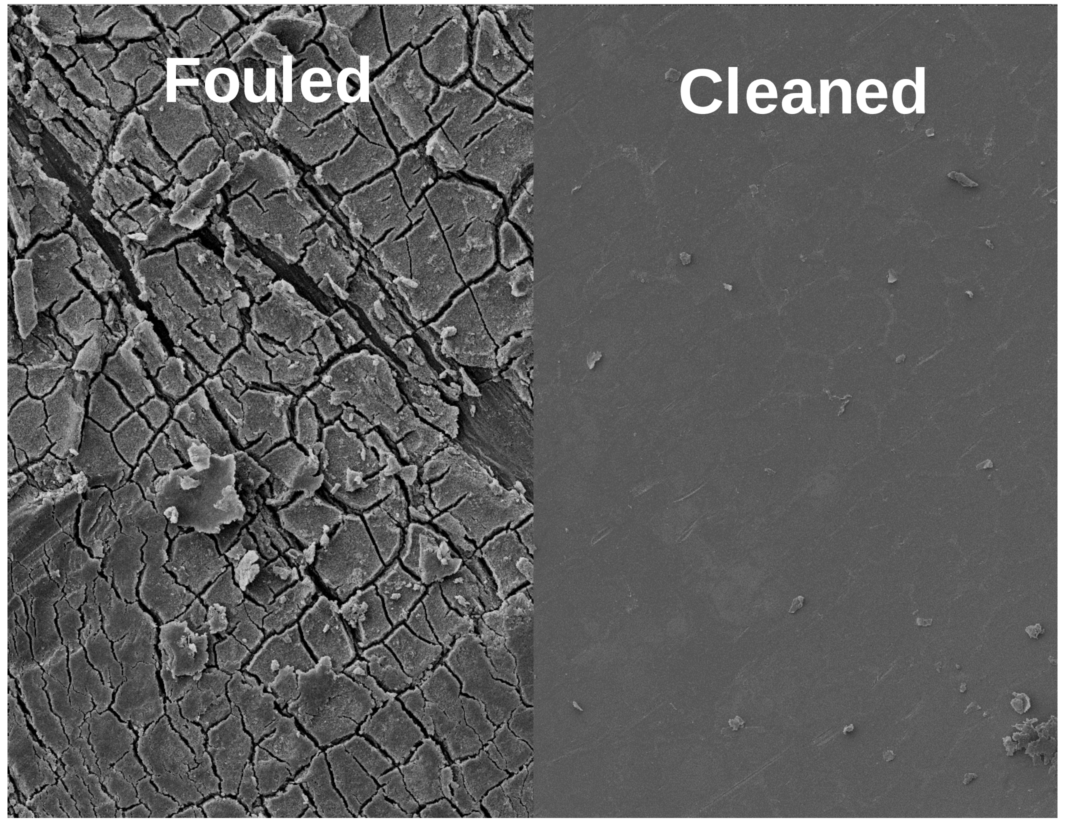
- Membrane processes
  - Advantages and challenges in food processing
  - Definition of membrane fouling
- Towards correlative X-Ray tomographic imaging
- Take home messages

Advantages of membrane processes for food processing

- Gentle product treatment due to moderate temperature changes during processing.
- High selectivity based on unique separation mechanism
  e.g. sieving, solution-diffusion or ion-exchange mechanism.
- Compact and modular design for ease of installation and extension.
- Low energy consumption compared to condensers and evaporators.
- Integration with other separation technologies in synergy or
  hybrid processes.

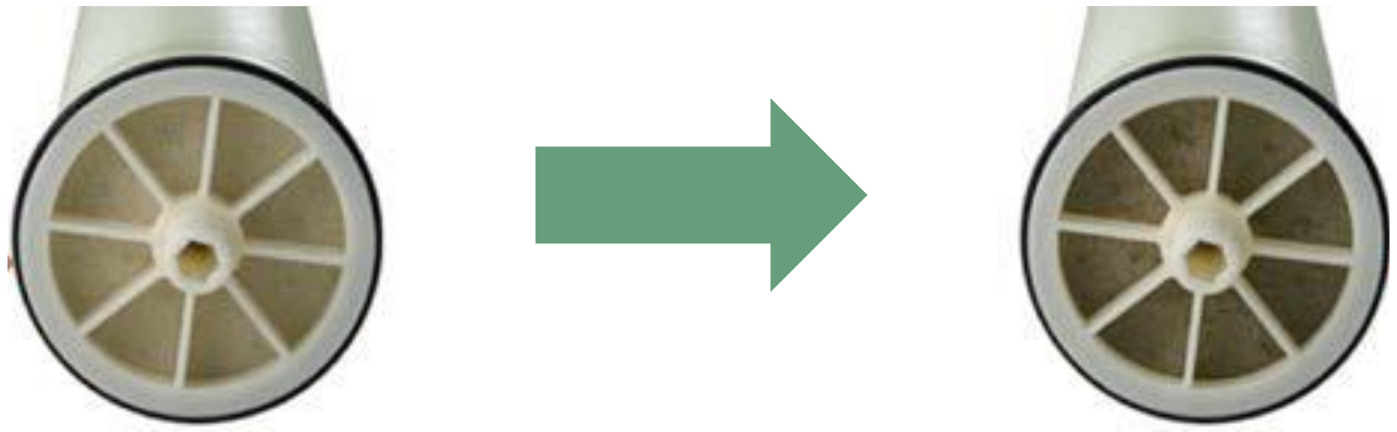
Challenges of membrane processes for food processing

- Short membrane life cycle and high membrane costs.
- Products have to be liquids or gaseous/vapour.
- Membrane performance is often limited by temperature, pH and chemical resistance.
- Limited economy of scale – membrane plants scale linearly.
- Tendency to **membrane fouling** and thus need for **membrane cleaning**.



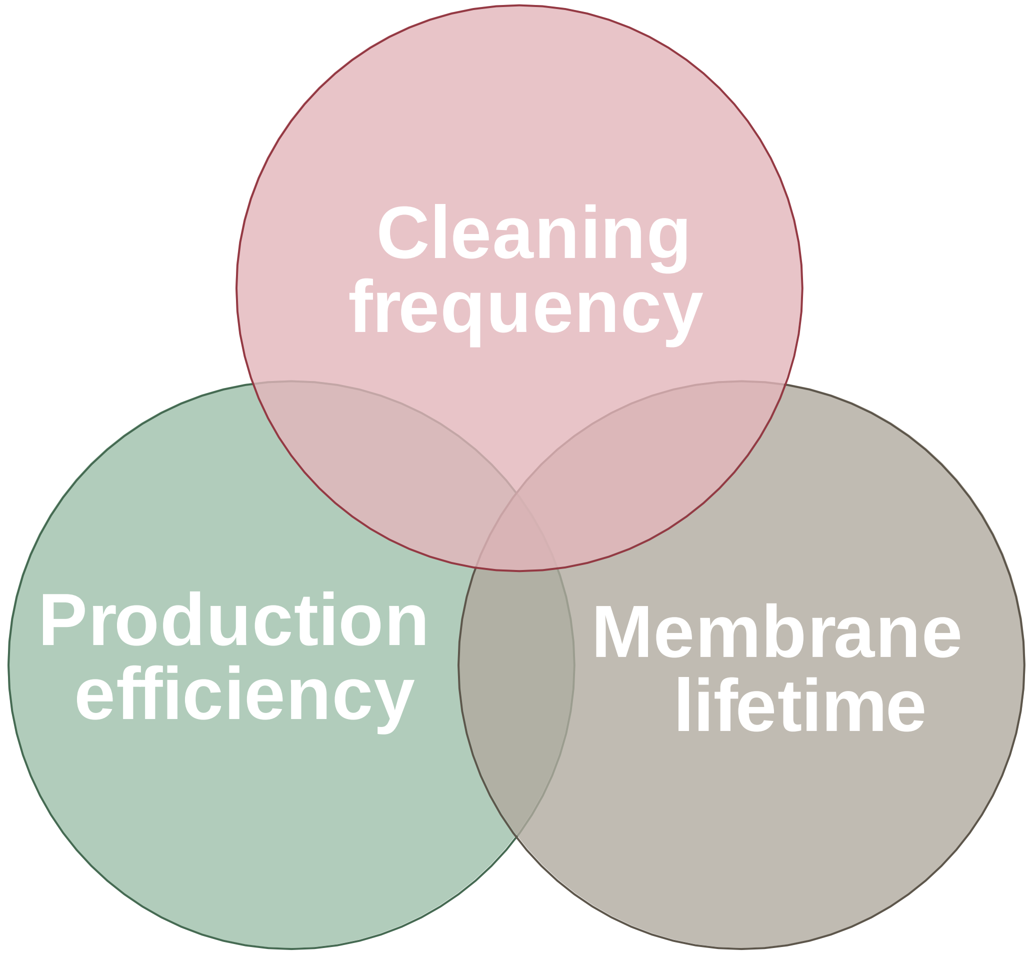
Membrane fouling?

“Process resulting in loss of performance of a membrane due to the deposition of suspended or dissolved substances on its external surfaces, at its pore opening, or within its pores”



Fouling and cleaning is costly

Production time losses due to cleaning and membrane replacement



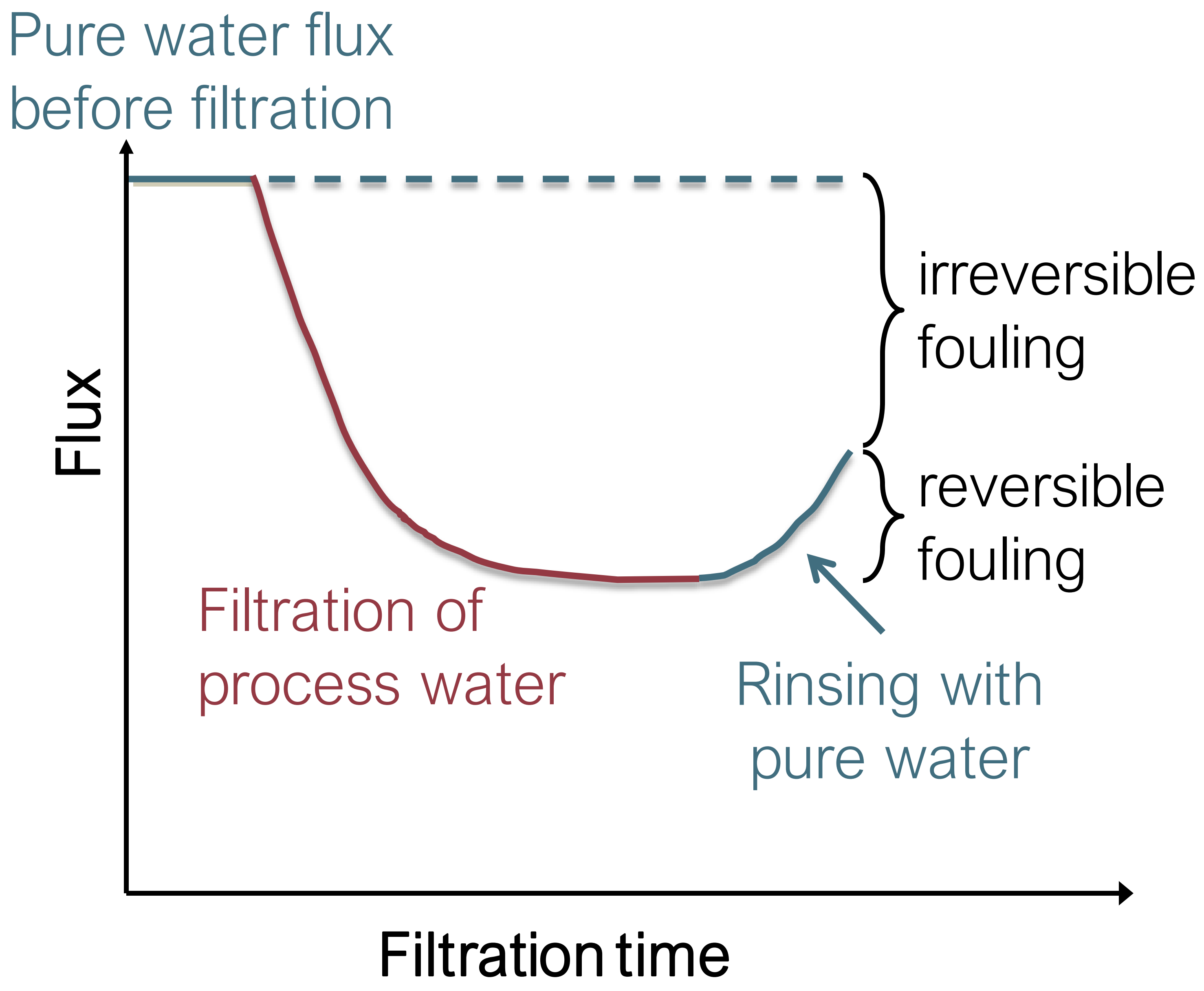
Irreversible fouling and cleaning cycles reduce membrane lifetime

Total daily plant capacity has to be increased as a consequence of cleaning

Annual costs

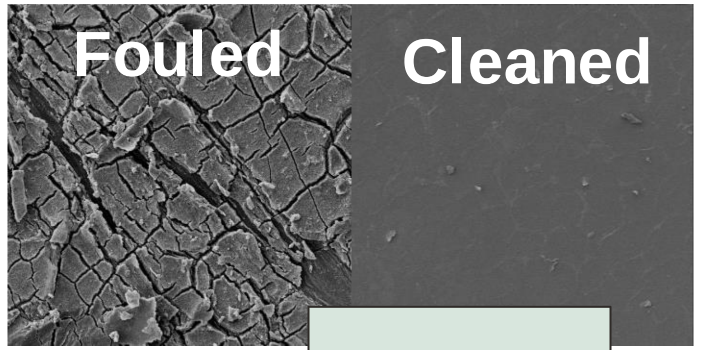
- Membrane cleaning: 5-20 % of CAPEX
- Membrane replacement: 2-5 % of CAPEX

Membrane fouling?

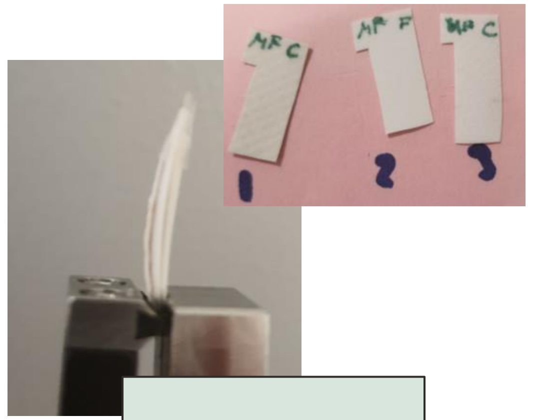


- No information about the fouling layer structure
- No information about membrane altering due to fouling and cleaning

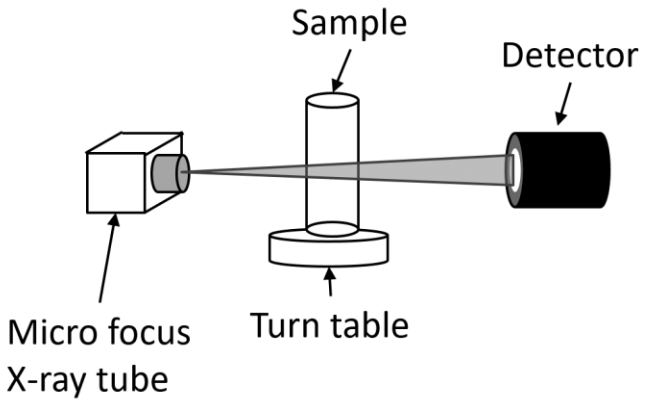
How we have worked with correlative X-ray tomogaphy



Fouling/ cleaning →



Sample mounting →



Scan with µCT →

Phase retrieval →

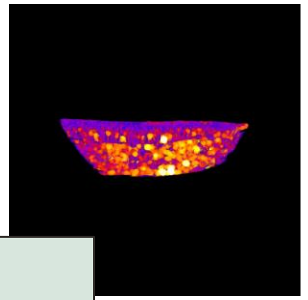
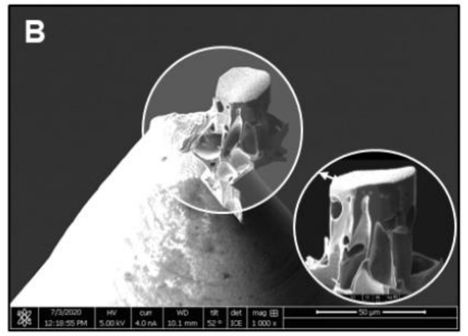


Image analysis

↓

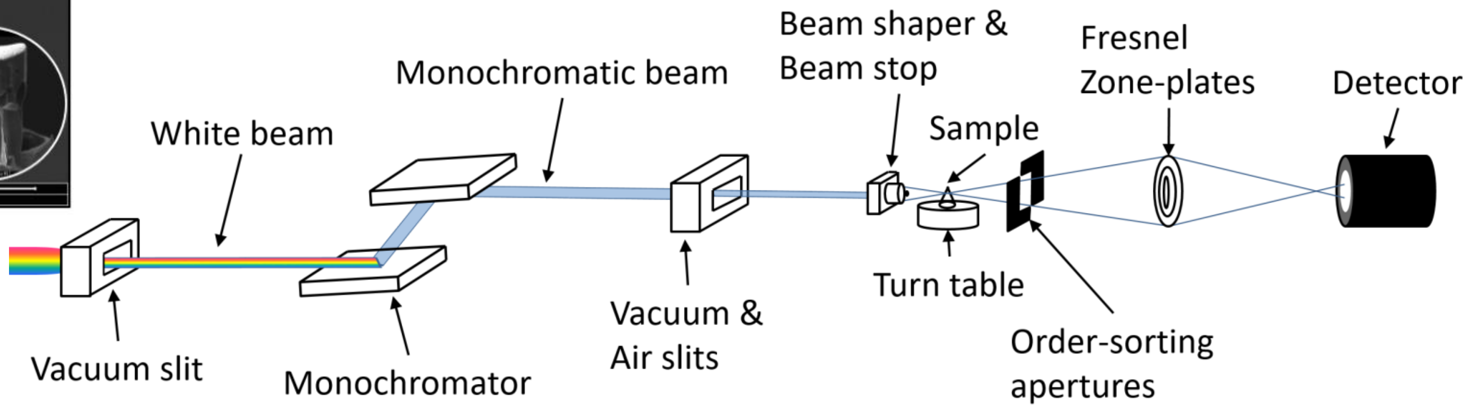
FIB:ing →



Scan with SRnCT

- *Higher contrast-to-noise ratio*
- *Higher resolution*
- *Higher flux => faster imaging, less dose on sample*

For beamtime proposal, important to motivate the need for SRnCT



<u>SRnCT including:</u>

- *Holotomography*
- *Full-field nanotomography*
- *Ptycho-tomography*

Toolkits for teaching tomography

RISE

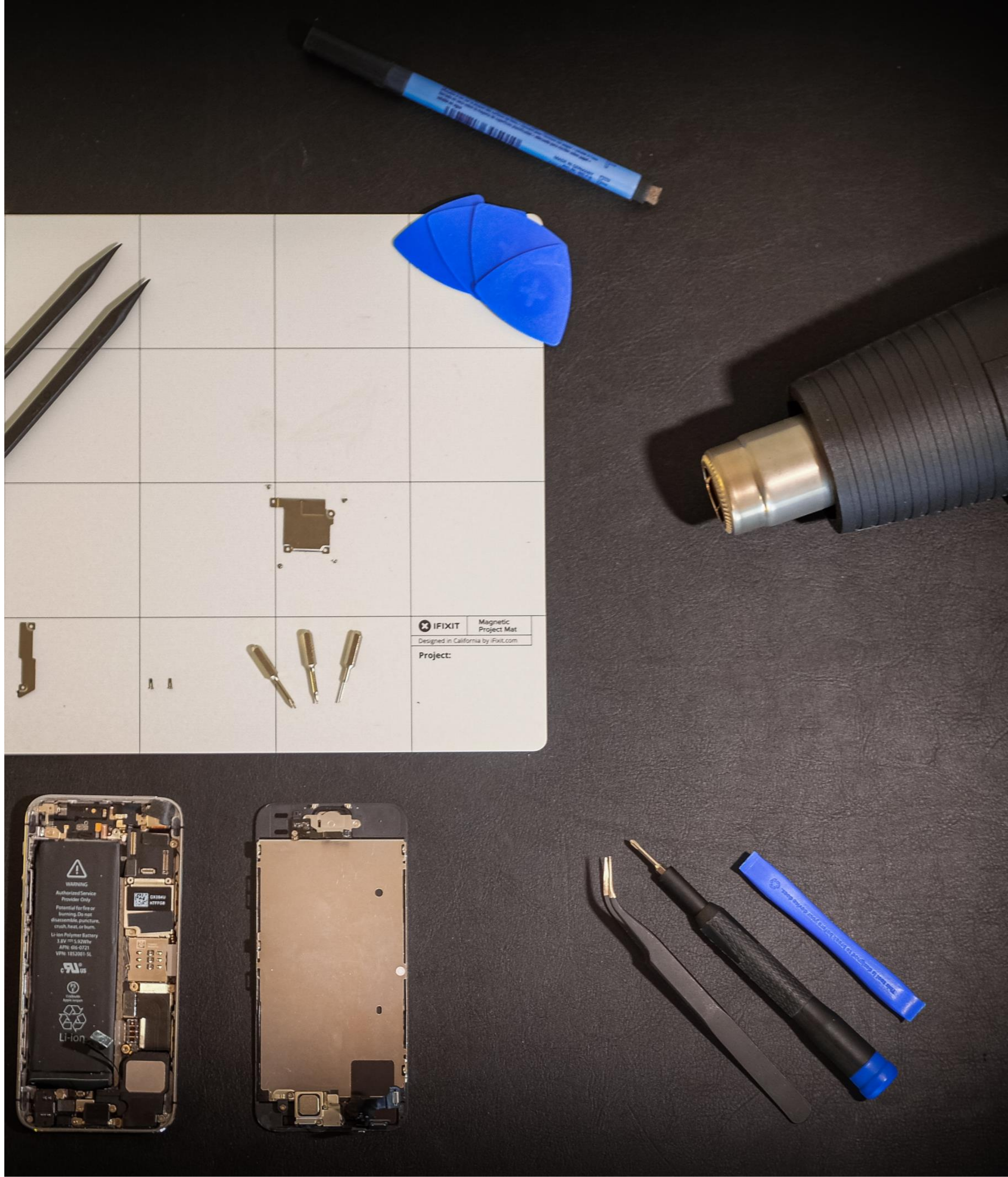
LUND UNIVERSITY

Additional lectures on:

- Small Angle X-ray Scattering (SAXS), Shun Yu, RISE
- High resolution coherent X-ray imaging, Pablo Villanueva-Perez, LU

VINNOVA
Sweden's Innovation Agency

Call: Increasing capacity and skills of PhD students regarding industrially relevant neutron and synchrotron-based analytical methods – 2019
Project: Measurements of membrane fouling in lignocellulosic biorefineries by ptycho-tomography, 2019-03613



Kitchen-Based Light Tomography – a home-based educational scanner for training the next generation of X-ray and Neutron tomography users

Emanuel Larsson[1] & Stephen A. Hall[1]
[1]Division of Solid Mechanics, Lund University, Lund, Sweden

ToScA Tomography for Scientific Advancement

LUND UNIVERSITY

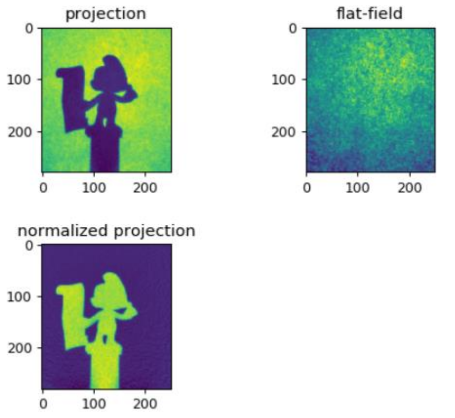
https://www.blue-scientific.com/tosca-2020/

Reconstruction – using TomoPy

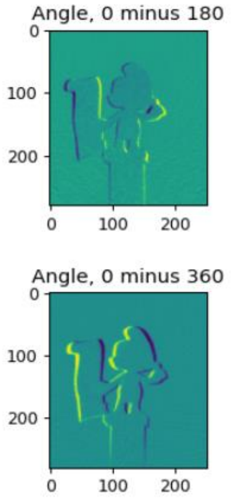
```
%matplotlib notebook

#plot figures
prj = plt.figure()
prj.subplots_adjust(hspace=.5)
ax = prj.add_subplot(2, 2, 1)
ax.set_title('projection')
ax.imshow(tomo[0])
ax2 = prj.add_subplot(2, 2, 2, sharex=ax, sharey=ax)
ax2.set_title('flat-field')
ax2.imshow(flat[0])
ax3 = prj.add_subplot(2, 2, 3, sharex=ax, sharey=ax)
ax3.set_title('normalized projection')
ax3.imshow(data[0])
plt.show()
```

Normalization



Tilt correction



3D Reconstruction

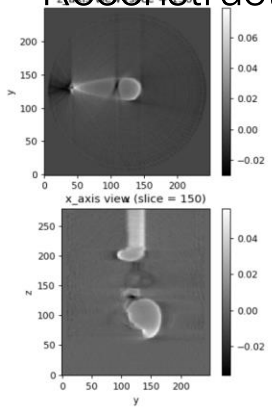
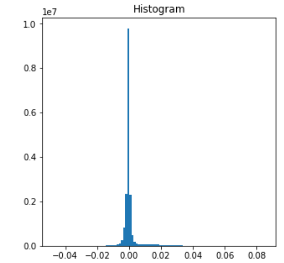


Image Processing and Quantitative Analysis

```
In [8]: # histogram of voxel values
        visualization.plot_histogram( slice, nbin=100 )
```

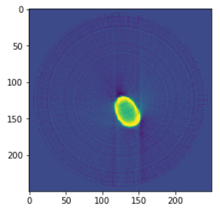


32 to 8 bit conversion

```
In [9]: #create new 32 bit stack
        slice_orig = slice

In [13]: #convert to 8-bit, 2D test slice
         l_val = -0.07
         u_val = 0.04

         test_slice = rescale_intensity(slice_orig[65], in_range=(l_val, u_val) )
         test_slice = img_as_ubyte(test_slice)
         imshow(test_slice)
         # histogram of voxel values
         visualization.plot_histogram( slice_orig[100], nbin=100 )
         visualization.plot_histogram( test_slice, nbin=100 )
```

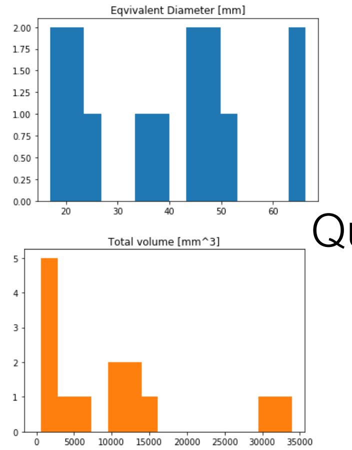


```
]: diameter = np.array([])
   tot_vol = np.array([])

   for i in range(no_inclus):
       diameter = np.append(diameter, props[i].equivalent_diameter)
       tot_vol = np.append(tot_vol, props[i].area * voxel_volume)

   parameter_list = [diameter, tot_vol]
   title_list = ['Eqvivalent Diameter [mm]', 'Total volume [mm^3]']

   for i in range(len(parameter_list)):
       plt.hist(parameter_list[i], bins=15)
       plt.title(title_list[i])
       plt.show(i)
```

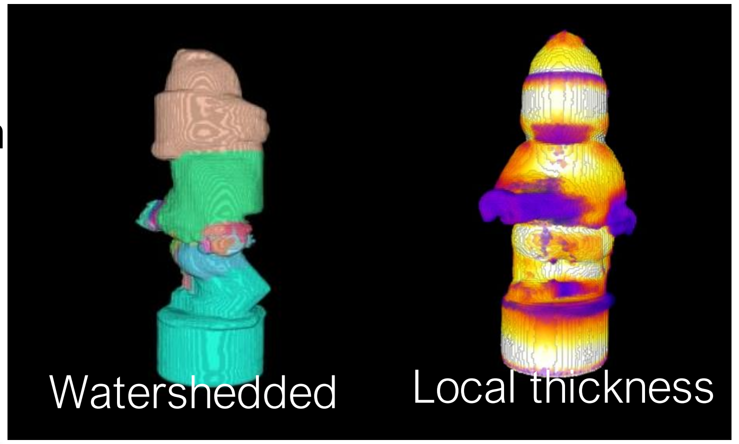


Quantitative analysis

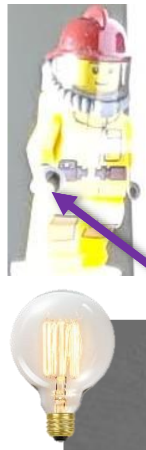
Conclusion:

"The obtained skills from developing/training in KBLT are directly transferable to real X-ray and Neutron tomography work."

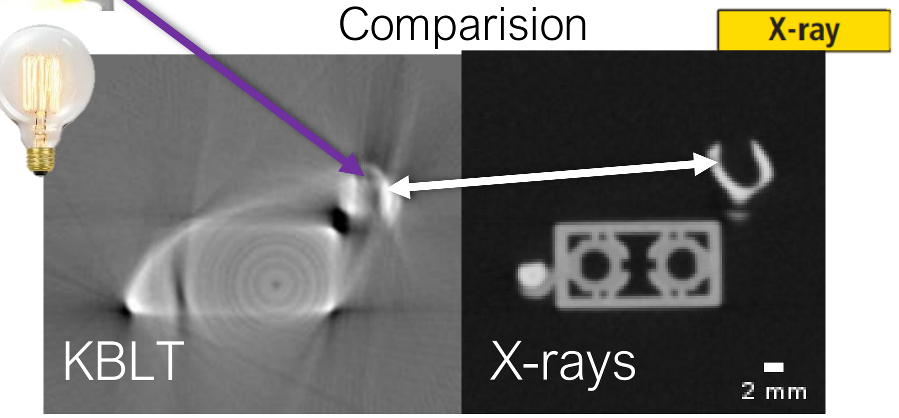
3D-rendering



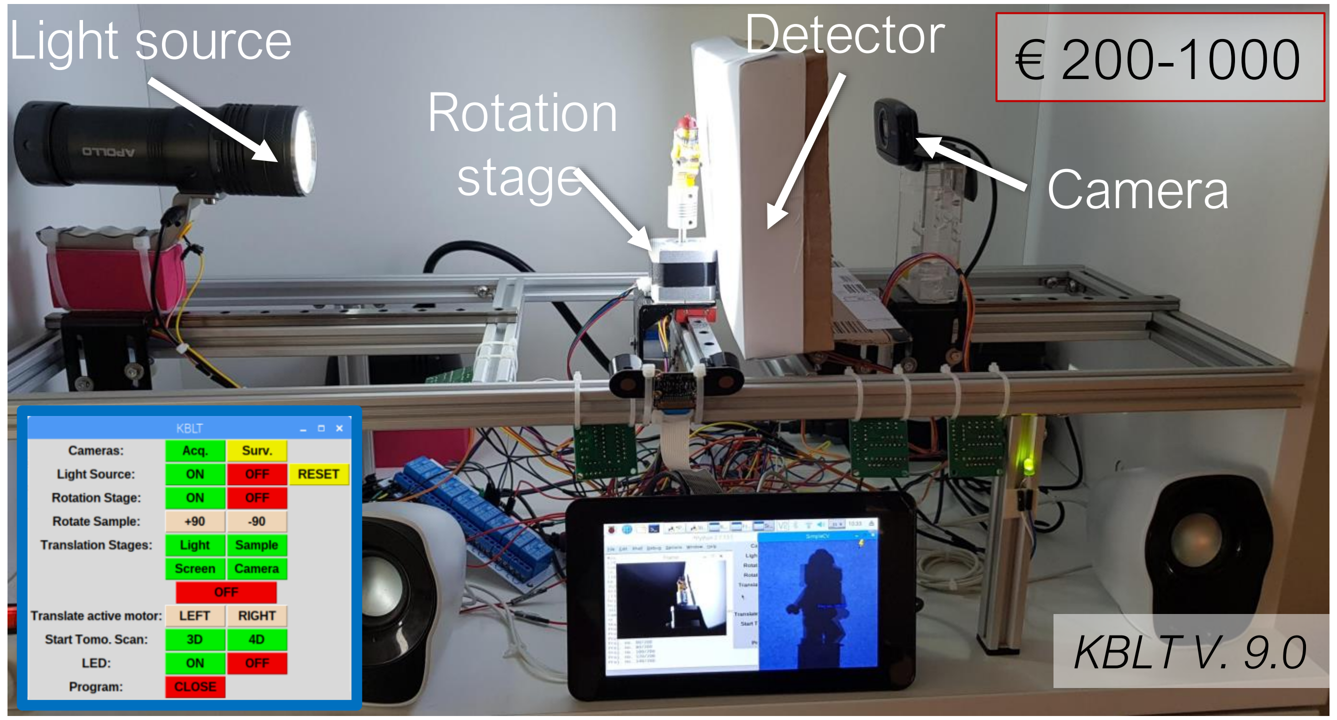
LegoMan

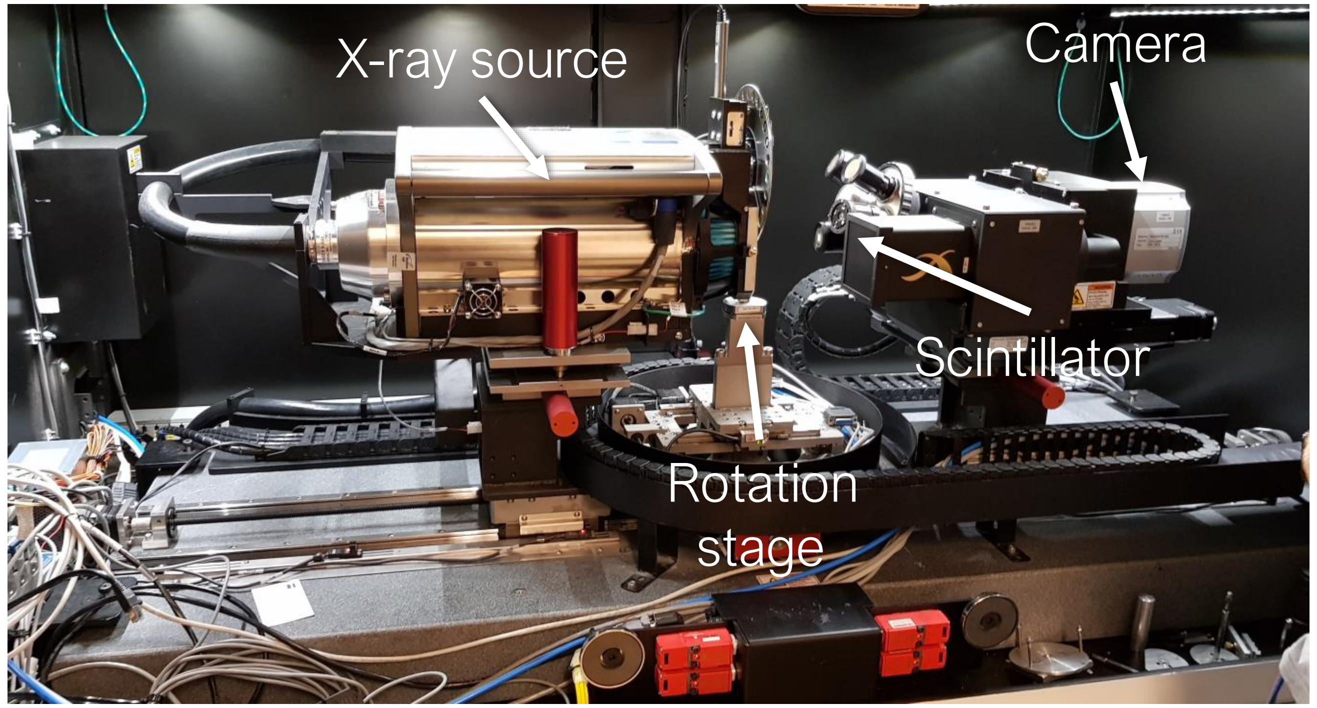


Comparision



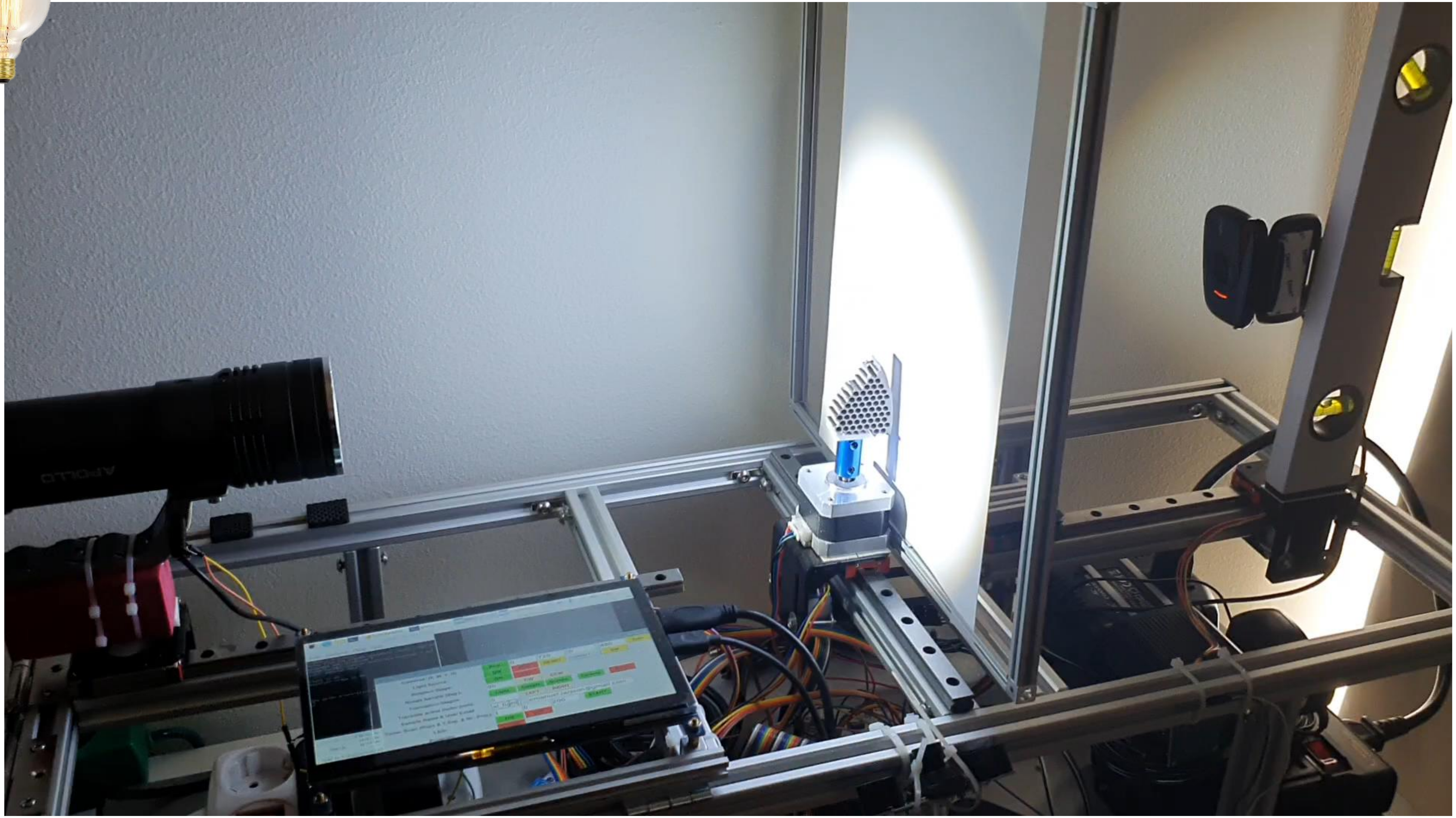
Has been used to train both PhD students and Post Docs.





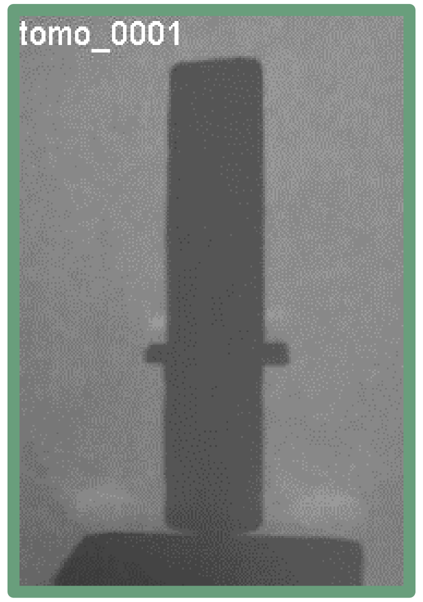
Zeiss Xradia 520 Versa at 4D Imaging Lab at Lund University.

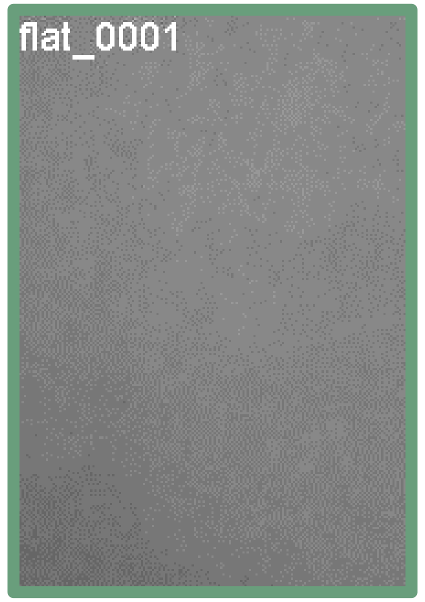
KBLT v.14.0 - pre-recorded demo!



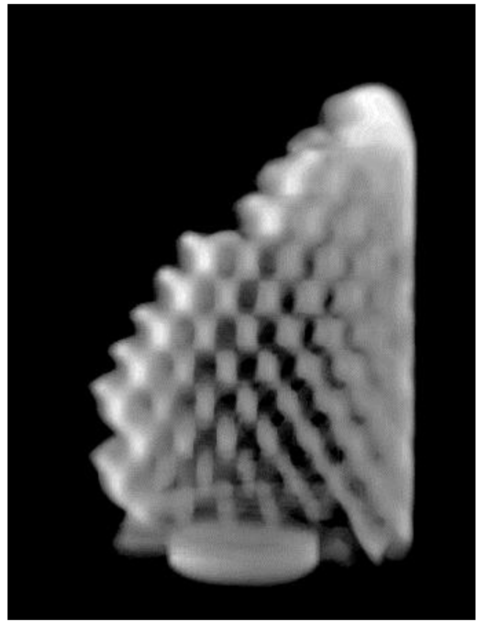
Membrane sample provided by Rudolph Gregor, Lund University, Sweden.

Camera view





3D-rendering



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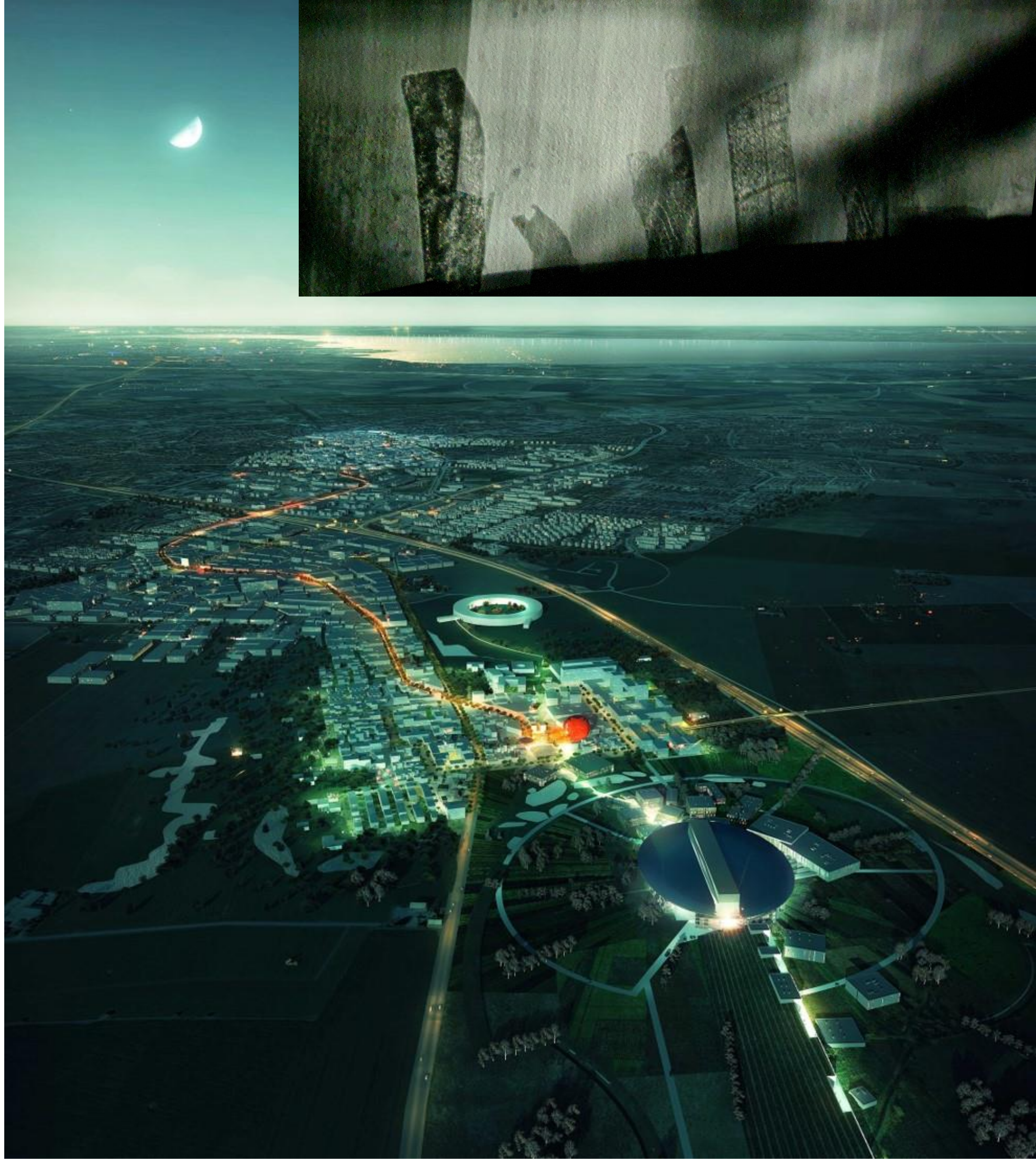
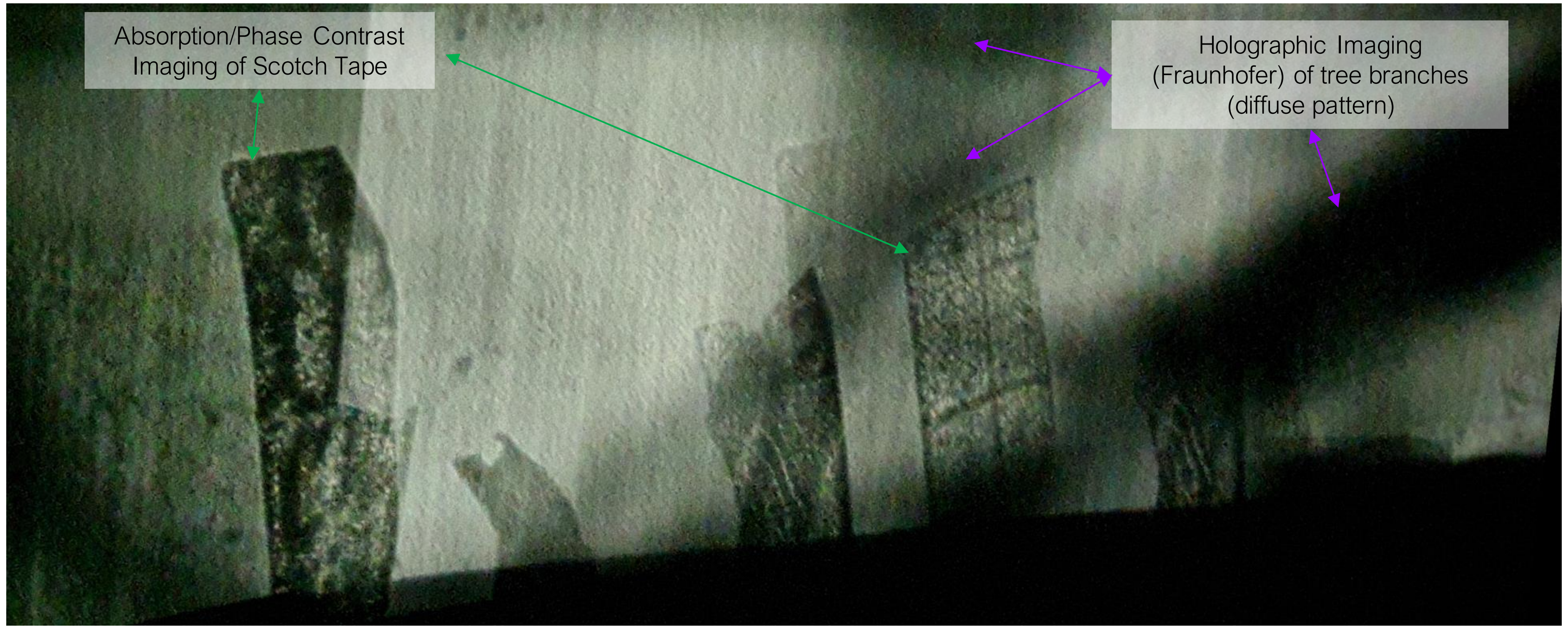
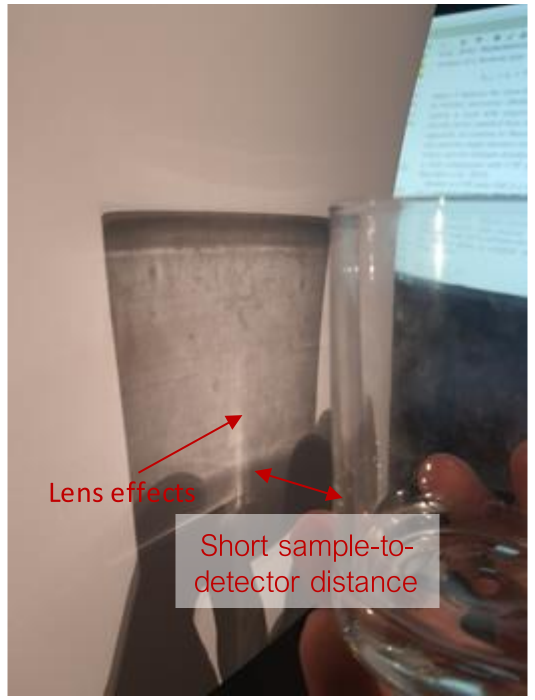
Imaging techniques with normal light vs. X-rays



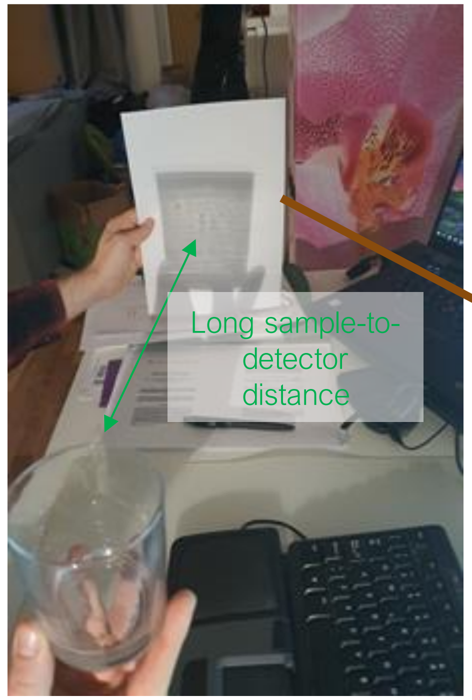


To reconstruction 'diffuse' patterns there is the need to perform so called 'phase retrieval', in order to decouple phase shift from absorption.

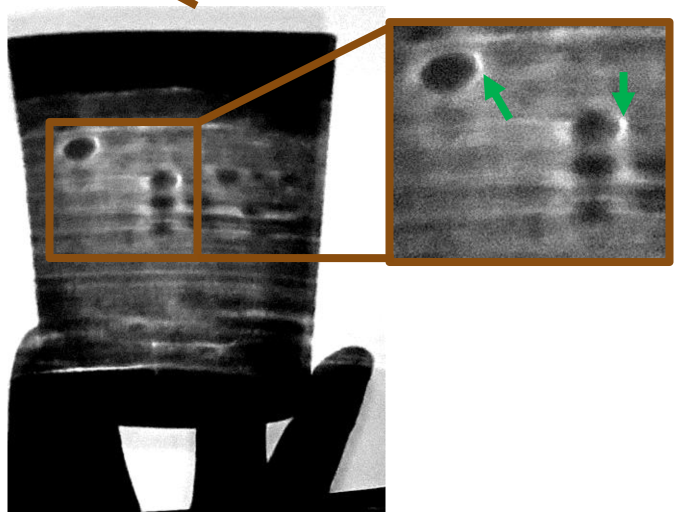
INCREASING RESOLUTION/
SAMPLE-TO-DETECTOR DISTANCE

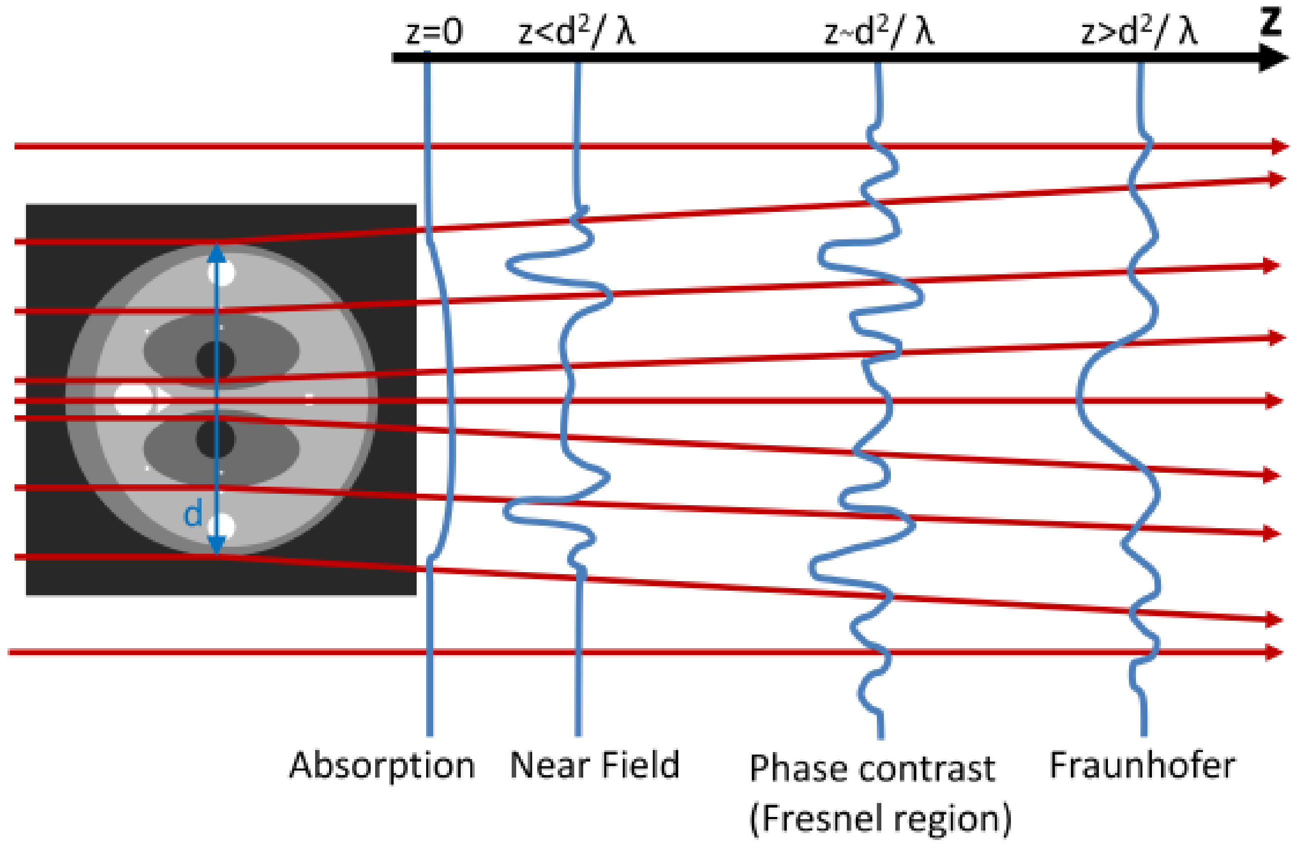


Absorbtion contrast



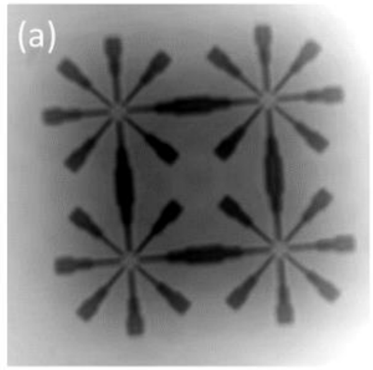
Phase contrast





X-ray

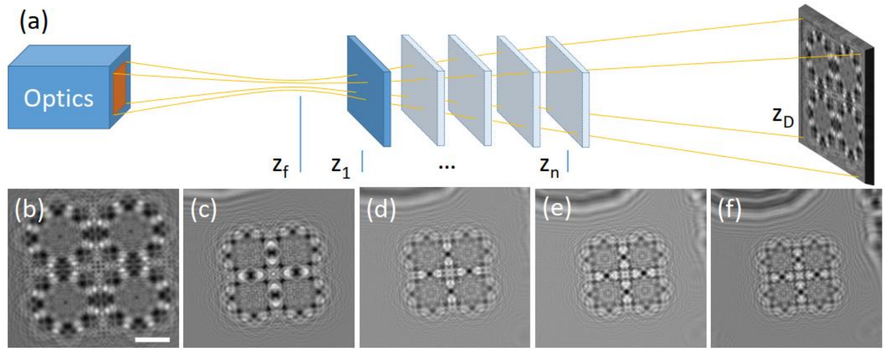
Full-field Synchrotron X-ray holographic nanotomography
- *Absorption*
- *Phase Contrast*
  1) Lensless
  2) Zone plate



Phase retrieved

2D test pattern

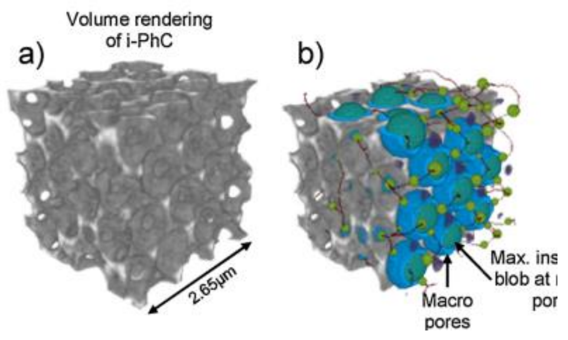
Typical Resolution: >100 nm



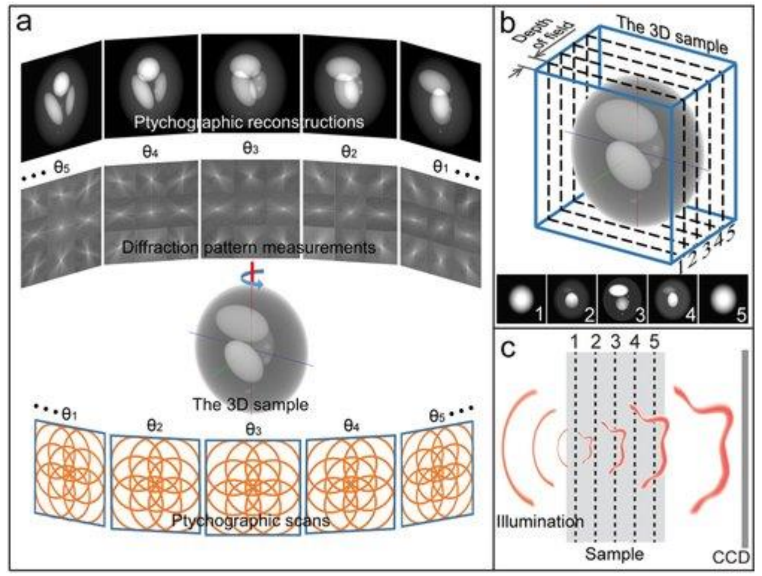
Langer, M., Zhang, Y., Figueirinhas, D., Forien, J. B., Mouton, C., Mokso, R., & Villanueva-Perez, P. (2020). PyPhase--a Python package for X-ray phase imaging. *arXiv preprint arXiv:2012.07942*.

Kalbfleisch S., Zhang Y., Kahnt M., Buakor K., Langer M., Dreier T., Dierks H., Stjarneblad P., Larsson E., Gordeyeva K.,Chayanun L., Soederberg D., Wallentin J., Bech M. and Villanueva-Perez P., X-ray in-line holography and holotomography at the NanoMAX beamline, *Submitted to the Journal of Synchrotron Radiation 2021.04.19*

Typical Resolution: >16 nm



Furlan, K. P., Larsson, E., Diaz, A., Holler, M., Krekeler, T., Ritter, M., ... & Janßen, R. (2018). Photonic materials for high-temperature applications: Synthesis and characterization by X-ray ptychographic tomography. *Applied materials today*, *13*, 359-369.

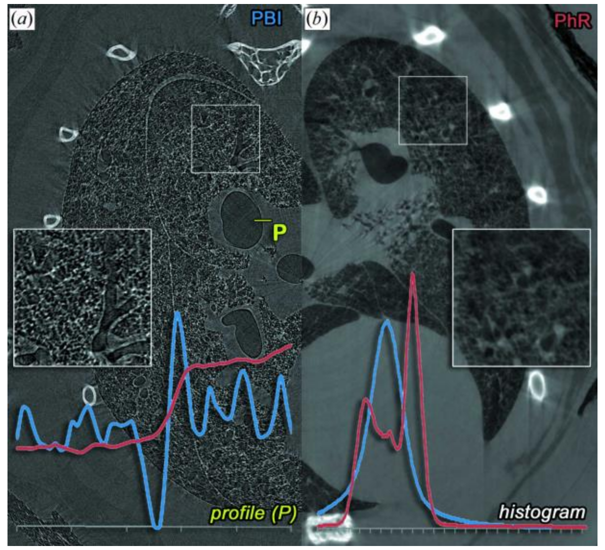


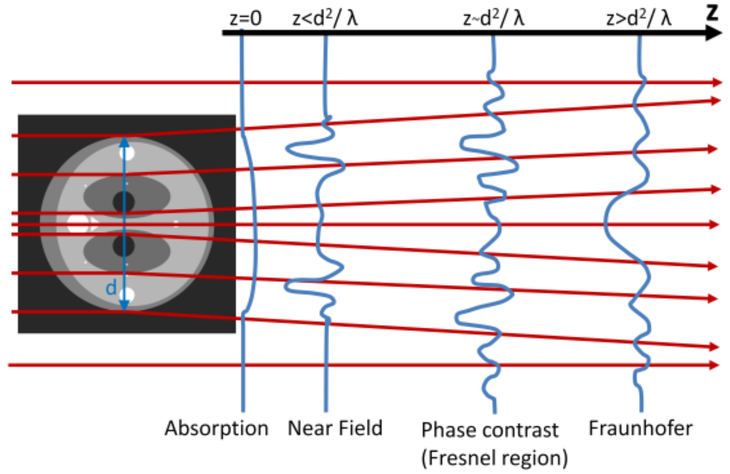
Li, P., & Maiden, A. (2018). Multi-slice ptychographic tomography. *Scientific reports*, *8*(1), 1-10.

Synchrotron X-ray ptycho-nanotomography
- *Absorption*
- *Phase Contrast*

INCREASING RESOLUTION/ SAMPLE-TO-DETECTOR DISTANCE

Full-field lab-based & Synchrotron X-ray Microtomography
- *Absorption*
- *In-line Phase Contrast*



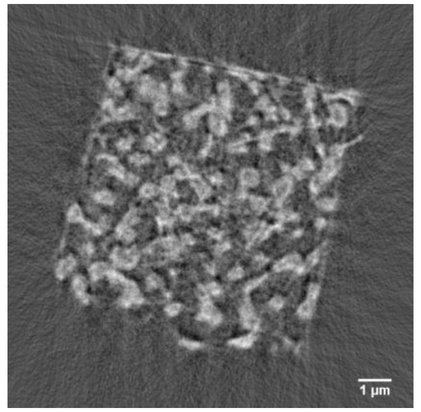


Mouse lung

Typical Resolution: >0.7 µm

Mohammadi, S., Larsson, E., Alves, F., Dal Monego, S., Biffi, S., Garrovo, C., ... & Dullin, C. (2014). Quantitative evaluation of a single-distance phase-retrieval method applied on in-line phase-contrast images of a mouse lung. Journal of synchrotron radiation, 21(4), 784-789.

Larsson, E., Gürsoy, D., De Carlo, F., Lilleodden, E., Storm, M., Wilde, F., ... & Greving, I. (2019). Nanoporous gold: a hierarchical and multiscale 3D test pattern for characterizing X-ray nano-tomography systems. *Journal of synchrotron radiation*, *26*(1), 194-204.

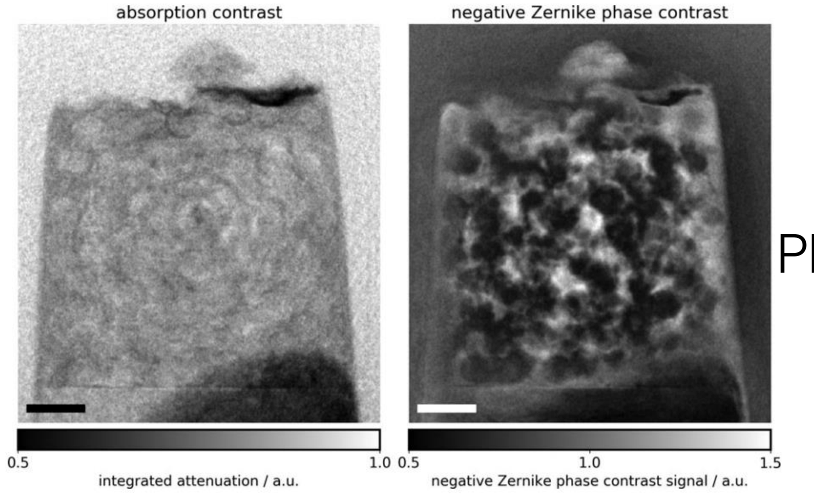


Nano-porous gold

Storm, M., Döring, F., Marathe, S., David, C., & Rau, C. (2020). The Diamond I13 full-field transmission X-ray microscope: a Zernike phase-contrast setup for material sciences. Powder Diffraction, 1-7.

Full-field Synchrotron X-ray nanotomography
- *Absorption*
- *Zernike Phase Contrast*

Typical Resolution: >60 nm



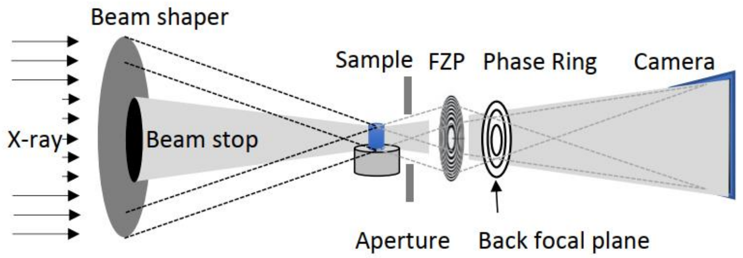
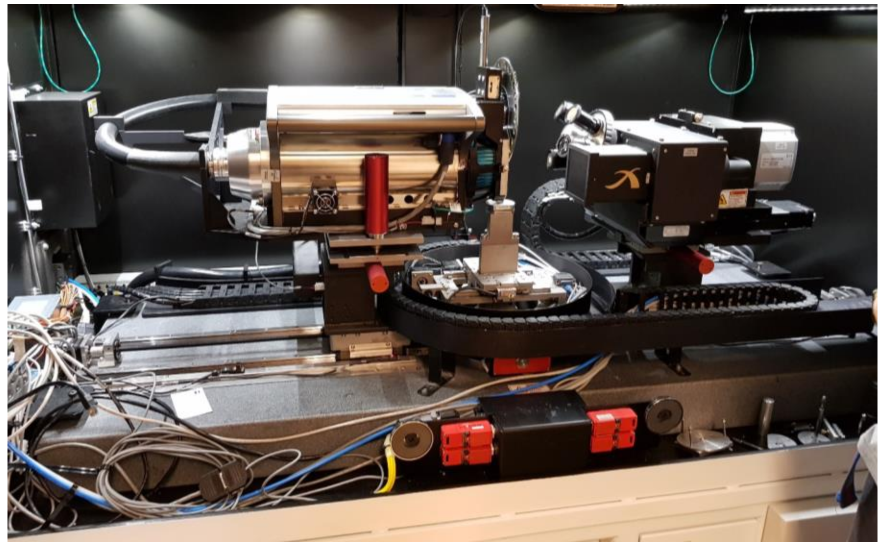


Figure. 1. Schematic of the ZPC setup at the imaging beamline P05.

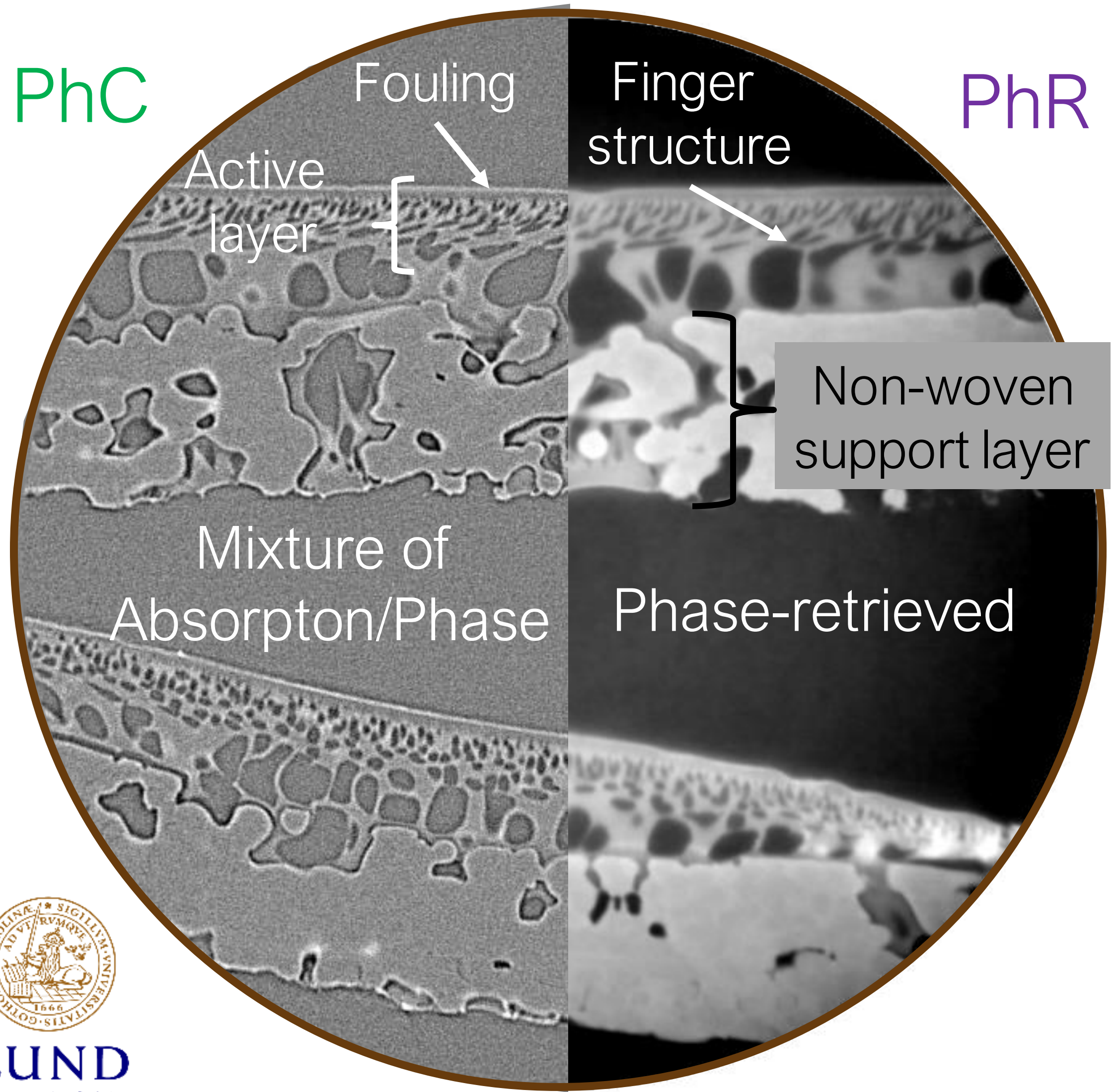
Flenner, S., Larsson, E., Furlan, K., Laipple, D., Storm, M., Wilde, F., ... & Greving, I. (2018). Nanotomography of inverse photonic crystals using zernike phase contrast. *Microscopy and microanalysis*, *24*(S2), 146-147.

Photonic glass

From microtomography towards ptycho-tomography of membrane fouling in lignocellulosic biorefineries



4D Imaging Lab, Division of Solid Mechanics, Faculty of Engineering, Lund University
https://www.solid.lth.se/resources/4d-imaging-lab/



Phase retrieval carried out by Frans Matsson & Rajmund Mokso using an in-house developed Python script.

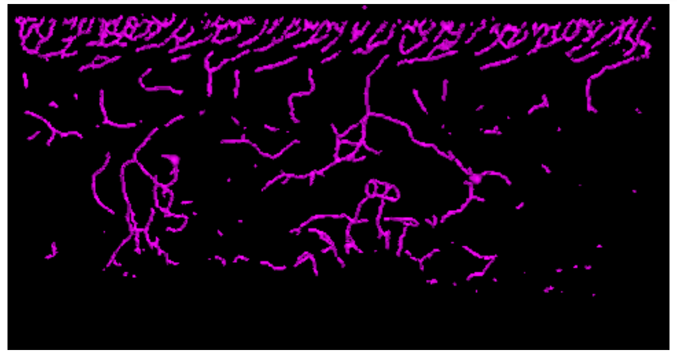
<u>Beamtime proposals sent in to:</u>

- NanoMAX, in total 2 during 2020-2021 - declined
- cSAXS, PSI, in total 2 during 2020-2021 (on waiting list)

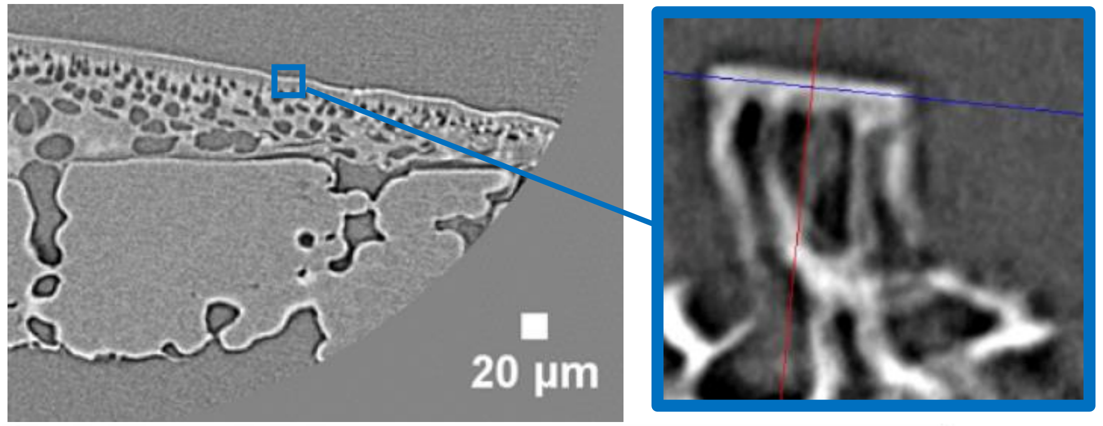
Center for Quantification of Imaging Data from MAX IV

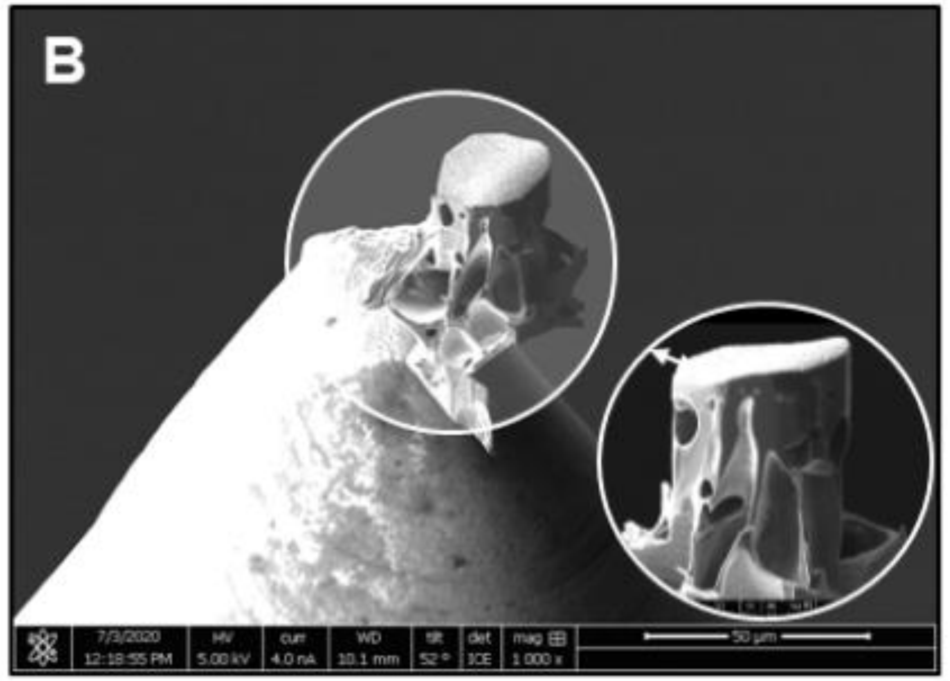
qimtools python package
https://qim.dk/

Image processing and analysis work is ongoing.



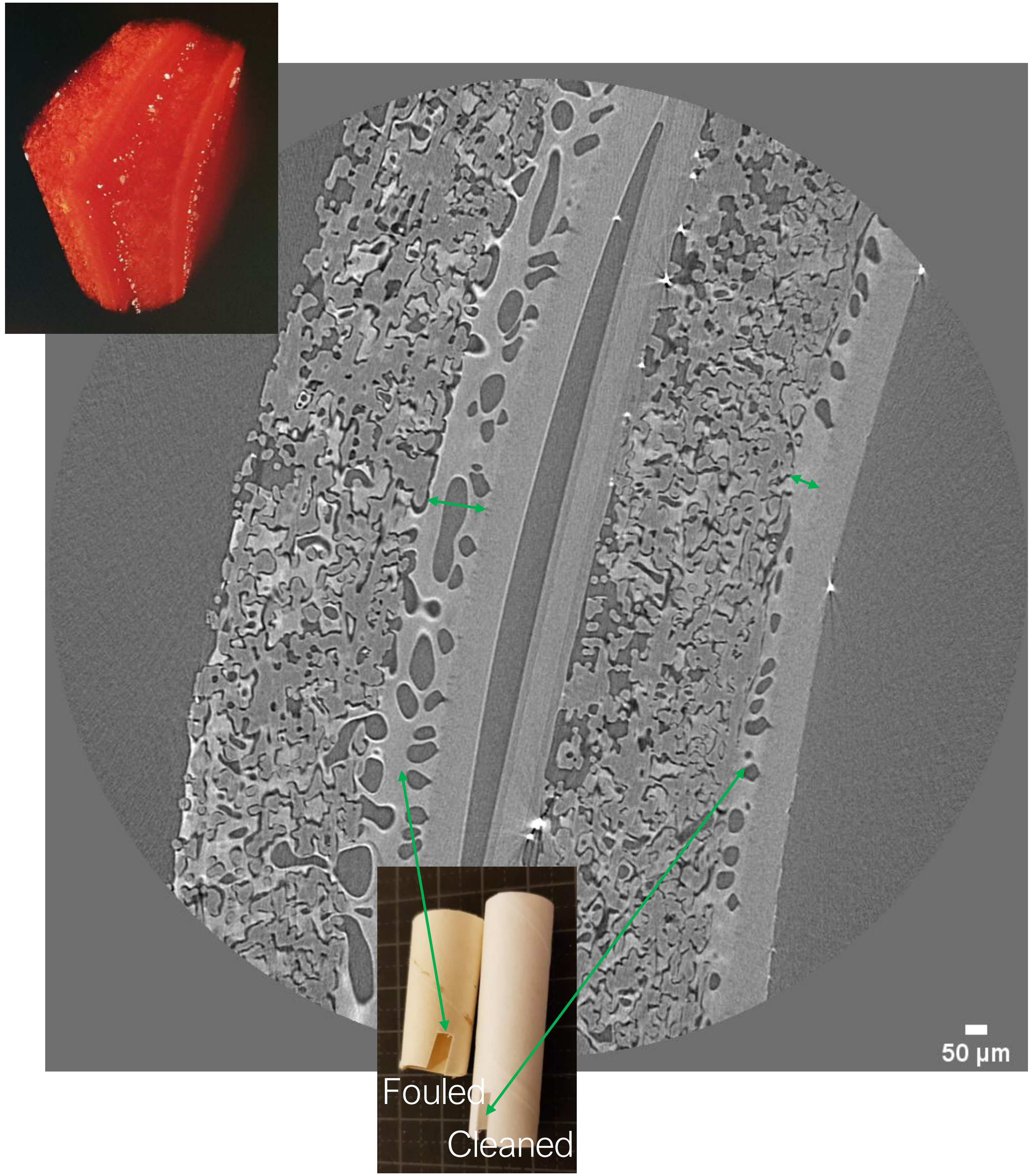
mCT scans of polysulfone-based membranes

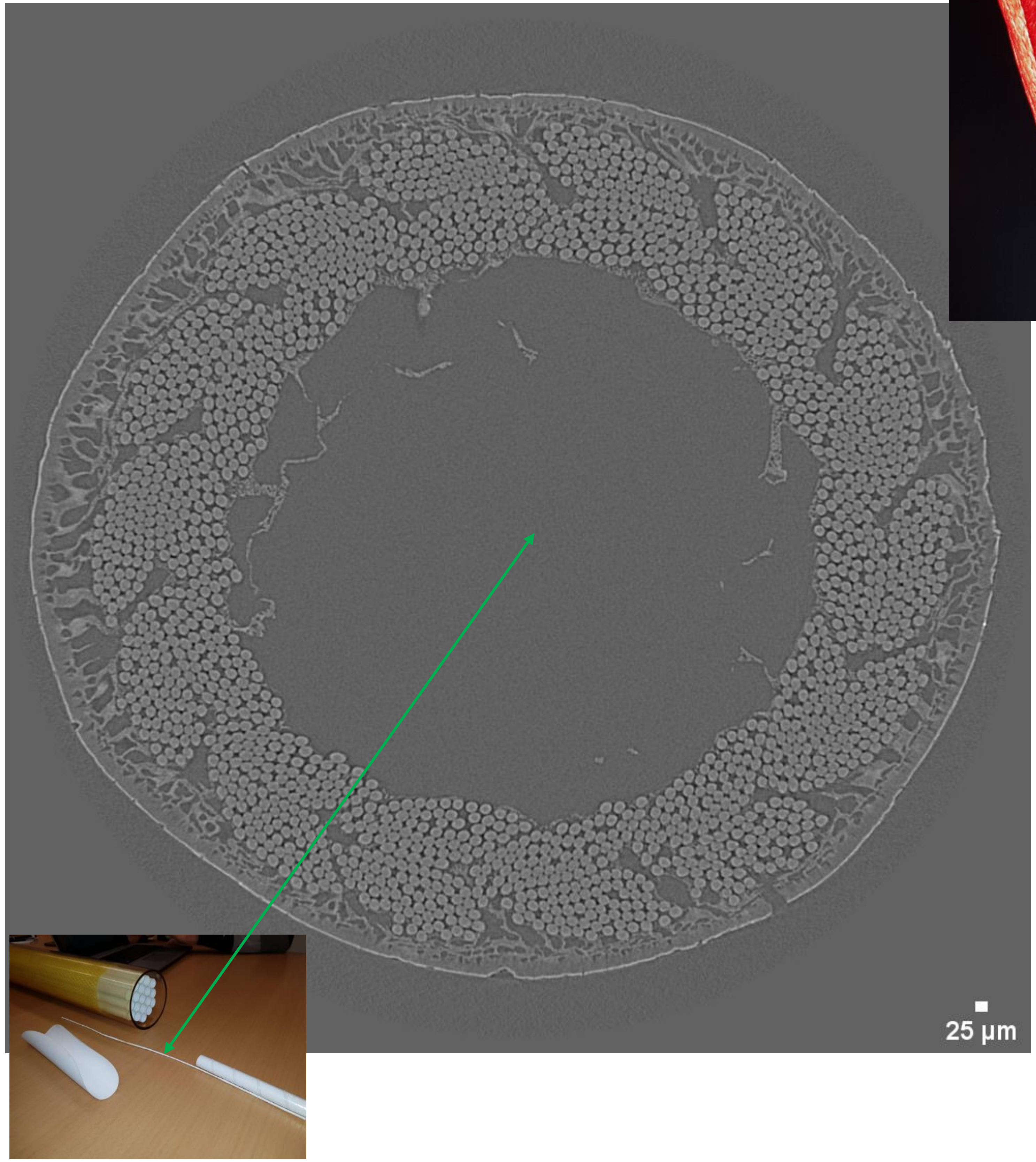




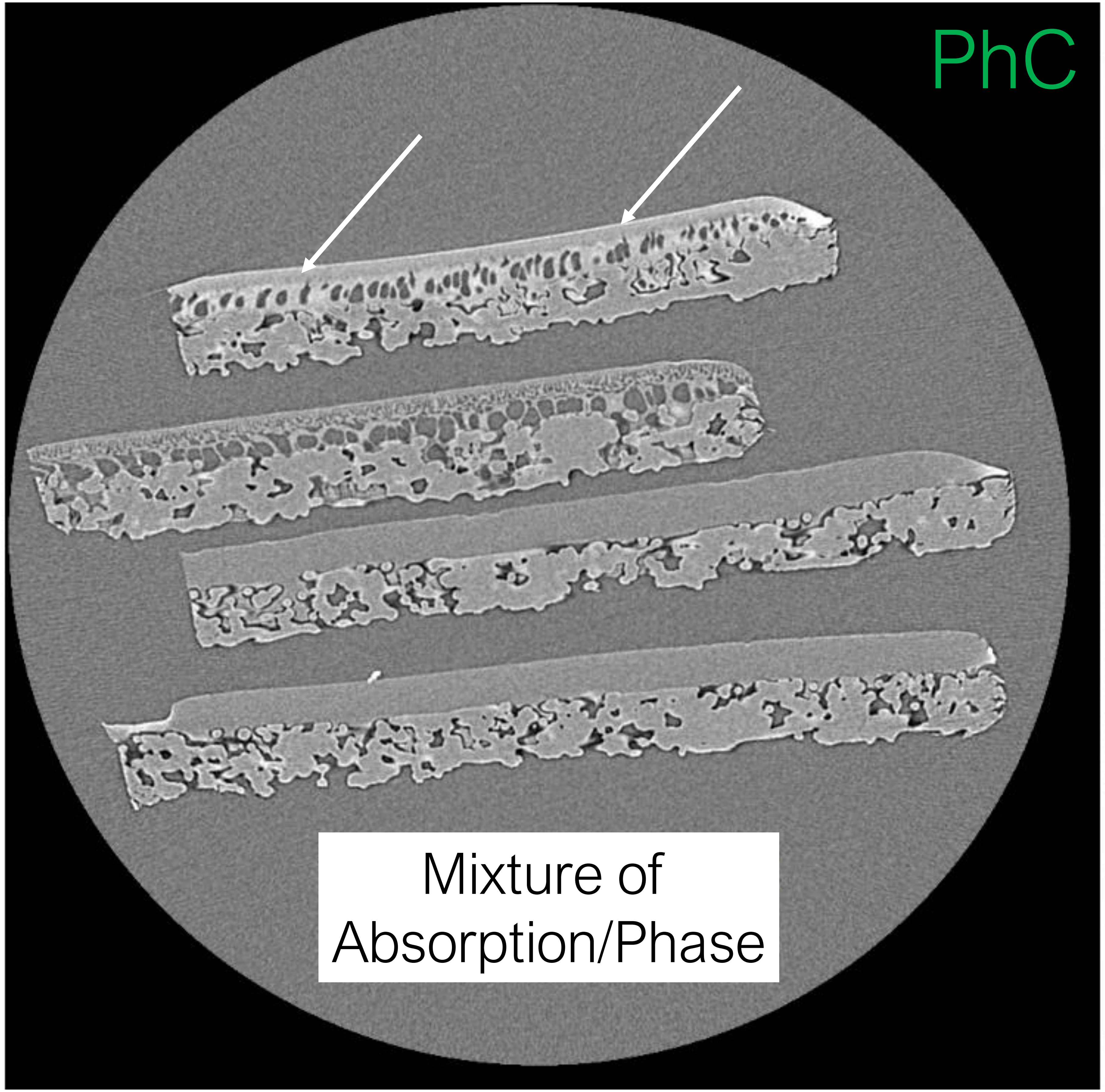
Sample preparation by FIB:ing (Focused Ion Beam) carried out by Torben Nilsson Pingel, RISE/Chalmers.

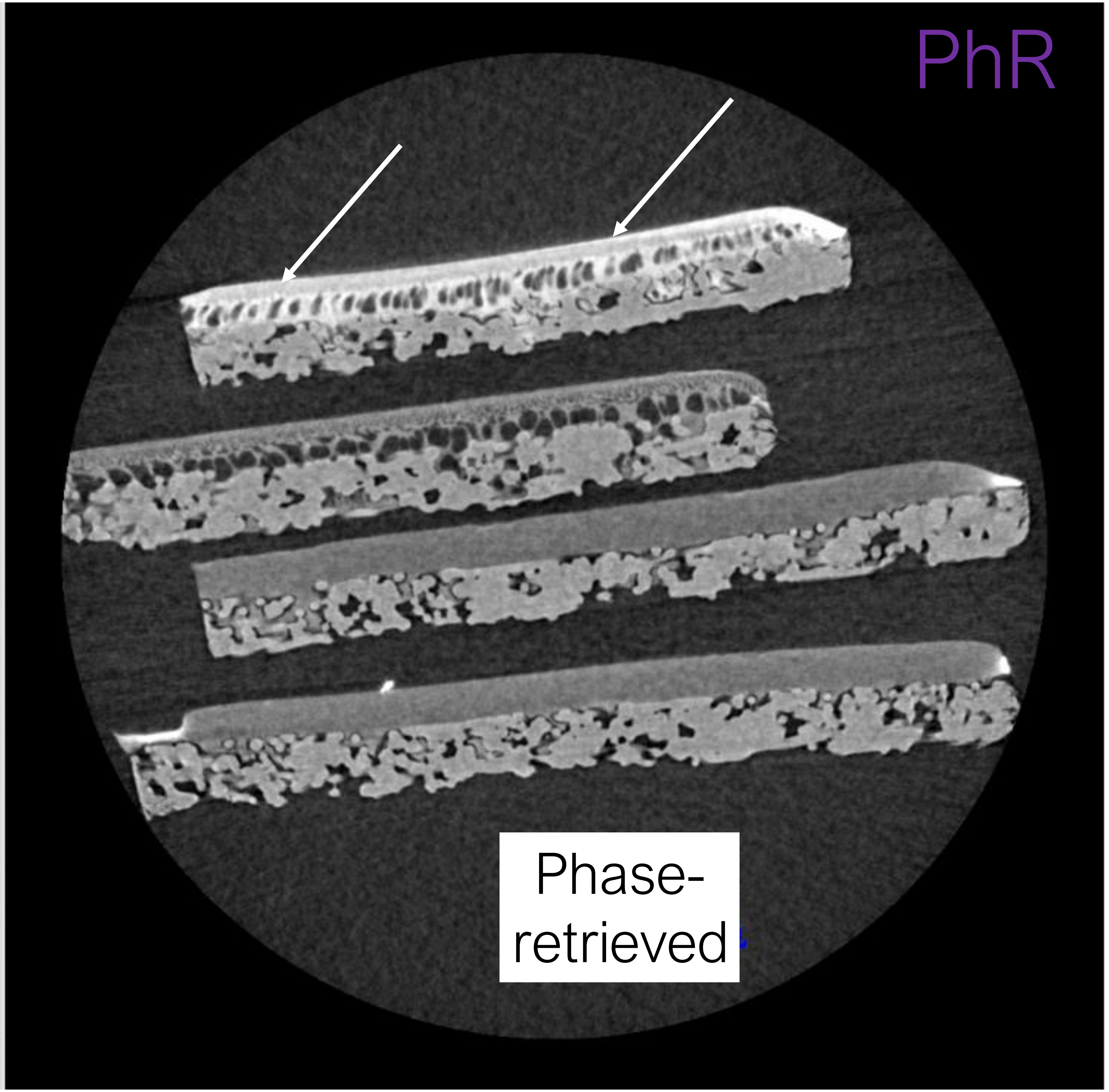
X-ray tomography of various membranes





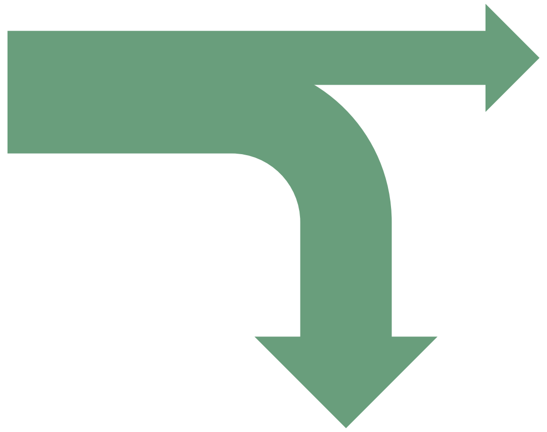
Non-phase retrieved vs phase retrieved





Rapeseed processing

3 kg
Seeds



1 kg
Oil

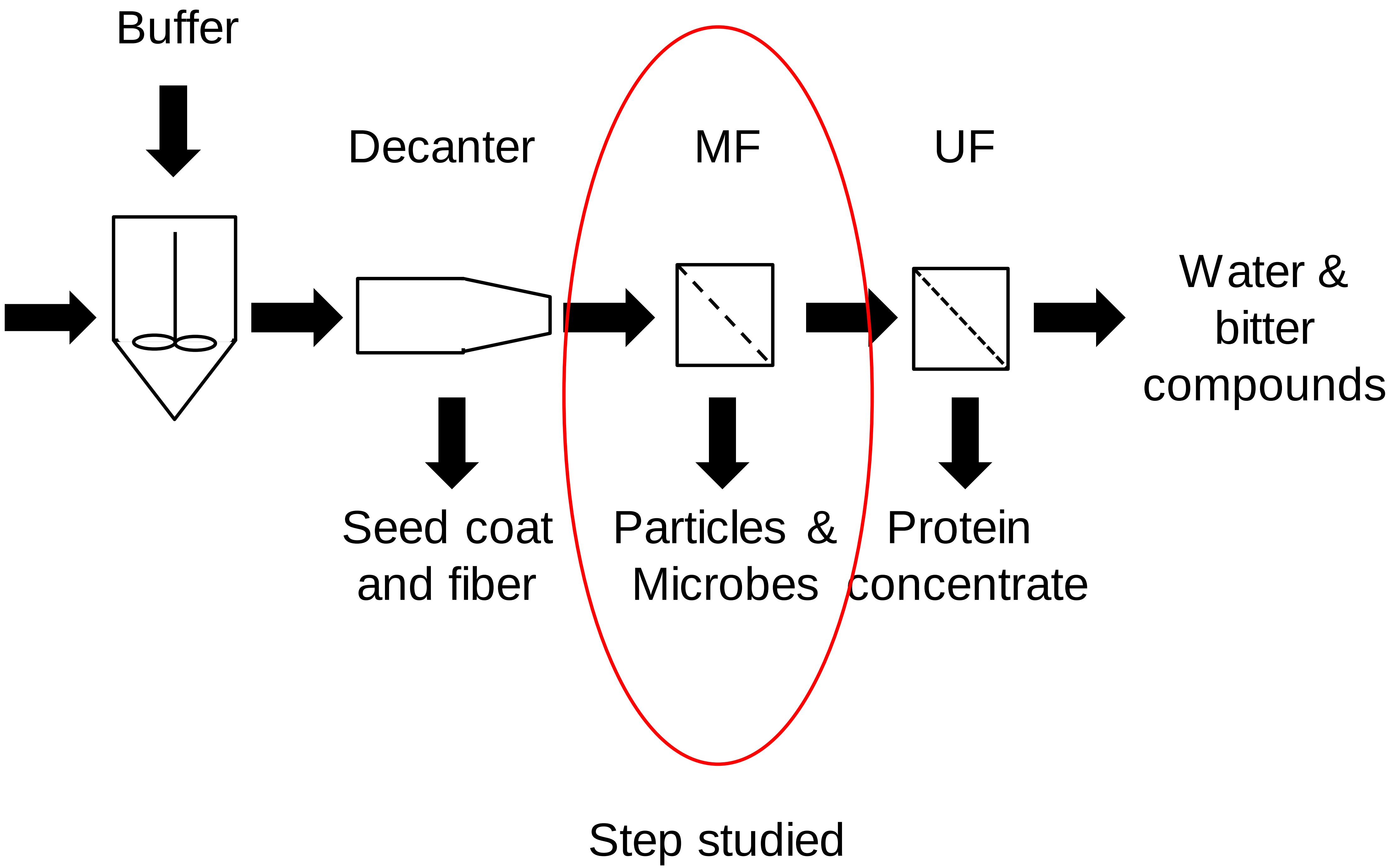
2 kg
Presscake
≈30 % Protein

Great amino acid composition

But bitter flavour

- World's second largest cultivated oilseed
- Rapeseed protein is comparable with soy protein in nutritional value
- Approved rapeseed protein isolated for human consumption in 2013 by European Food and Safety Authority

Rapeseed processing



Lab µCT scans of fouled and cleaned MF membranes

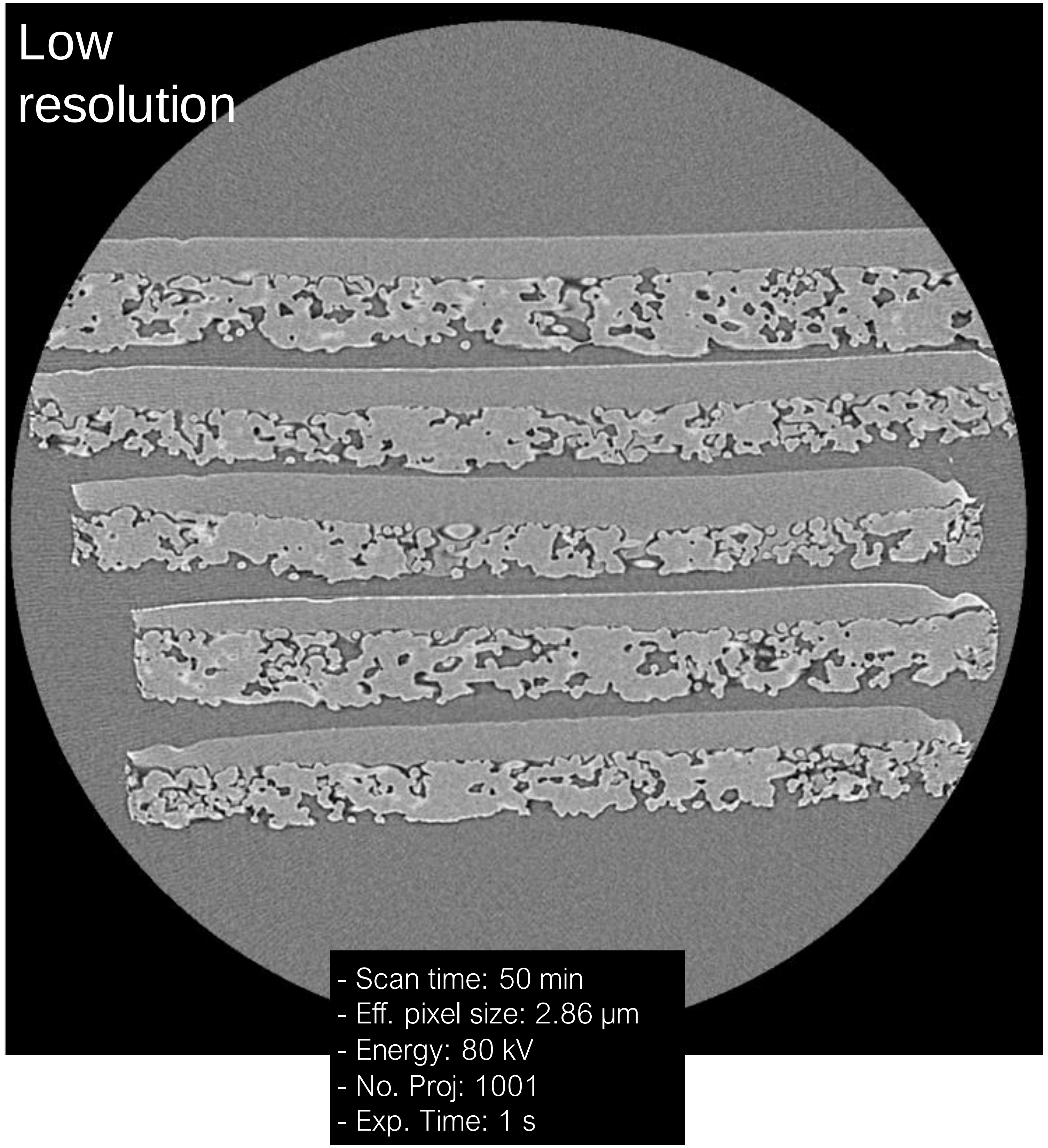
MFG2 Pristine

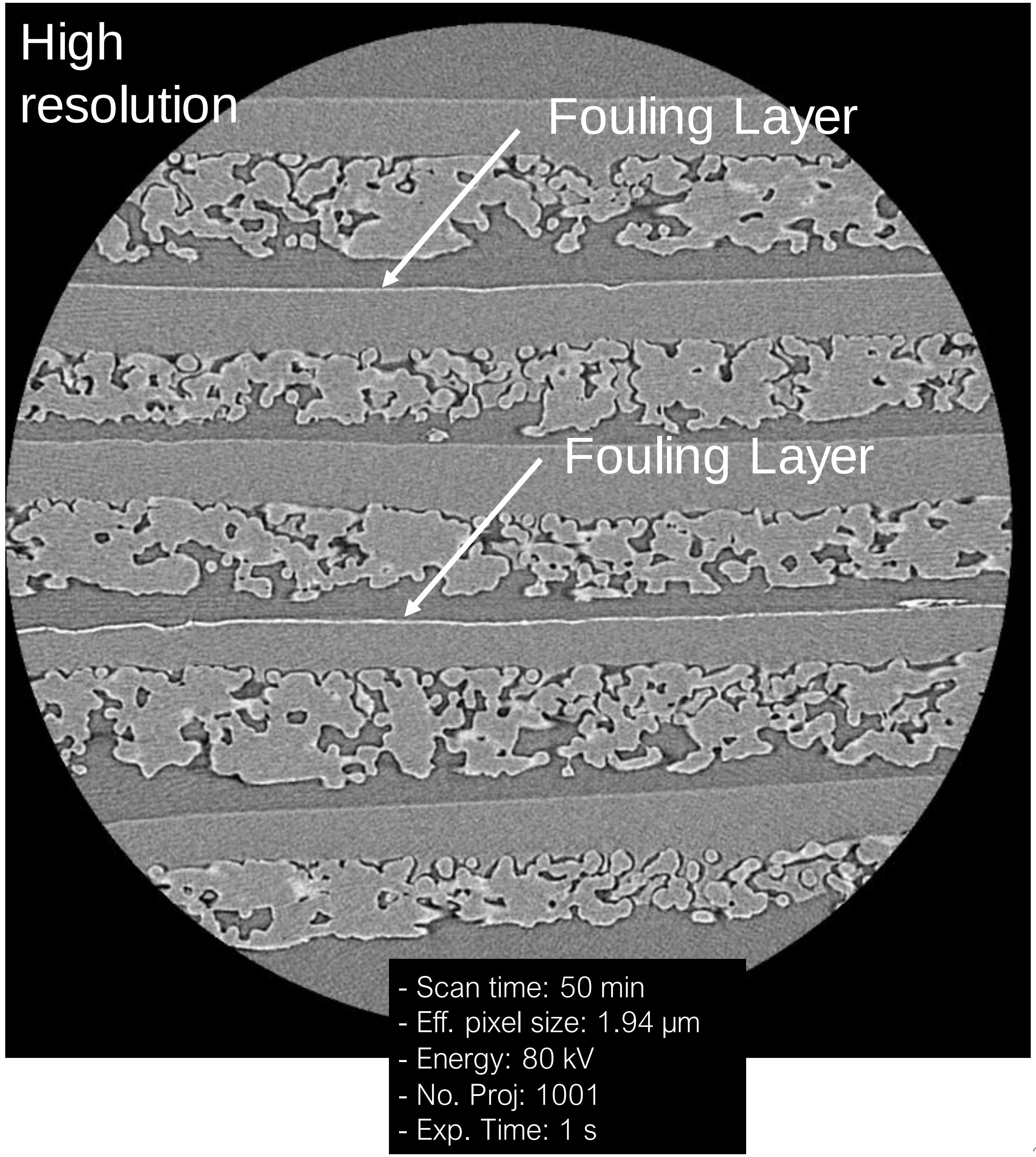
MFG2 Fouled

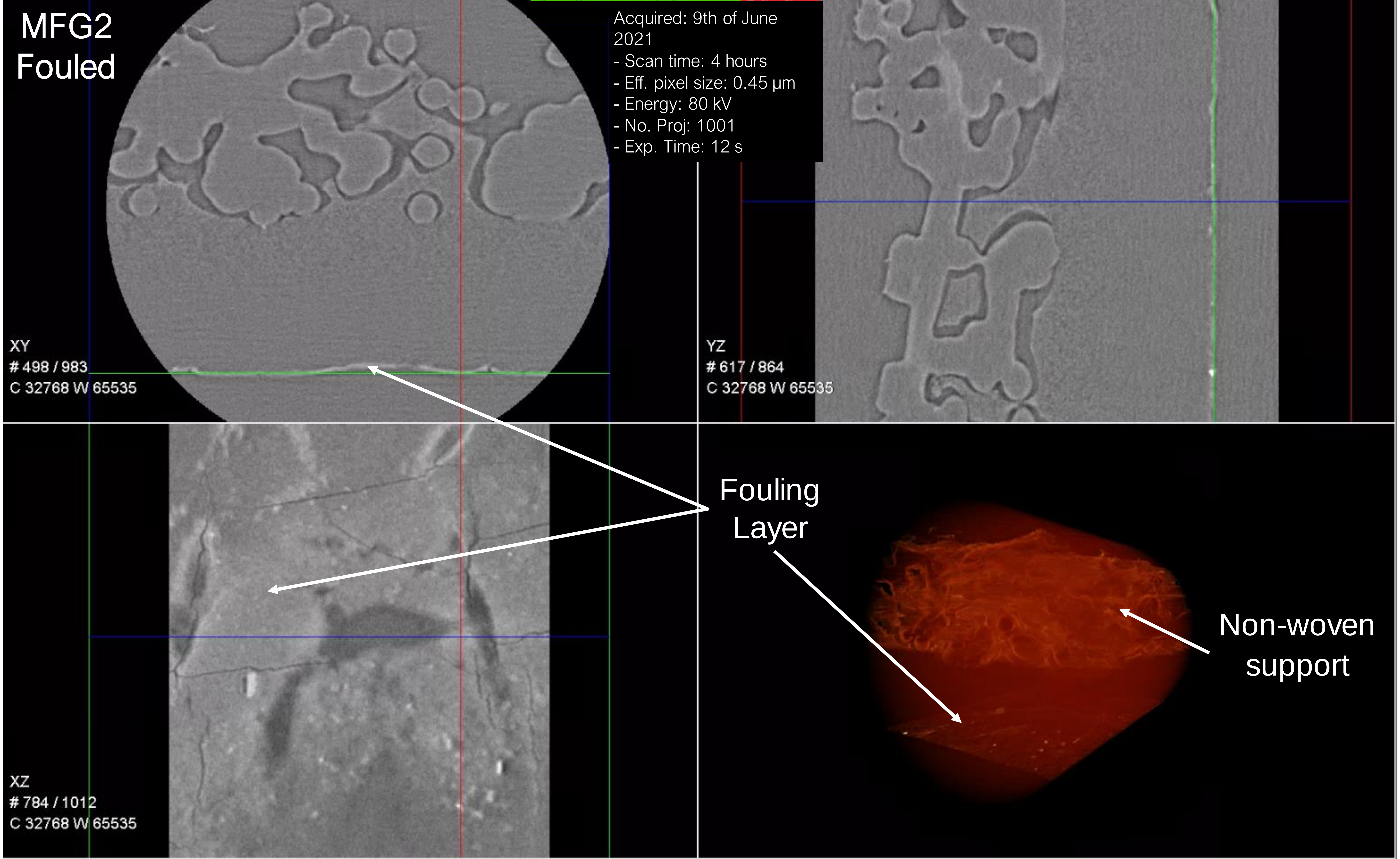
MFG2 Cleaned

MFG1 Fouled

MFG1 Cleaned






MFG2
Fouled
Acquired: 9th of June 2021
- Scan time: 4 hours
- Eff. pixel size: 0.45 µm
- Energy: 80 kV
- No. Proj: 1001
- Exp. Time: 12 s
XY
498 / 983
C 32768 W 65535
YZ
617 / 864
C 32768 W 65535
XZ
784 / 1012
C 32768 W 65535
Fouling
Layer
Non-woven
support

Take home messages

- X-ray tomographic imaging provides novel insight into the impact of membrane fouling and cleaning on the membrane structure

- High resolution available at synchrotron facilities is needed for detailed analysis

- The technology requires a wide network of experts and collaboration

Thank you for listening!

Dr. Shun Yu
Dr. Torben Nilsson Pingel

Stiftelsen Lantbruksforskning

Mr. Gregor Rudolph
Dr. Johan Thuvander
Dr. Emanuel Larsson
Assoc. Prof Stephen Hall
Assoc. Prof. Pablo Villanueva-Perez
Prof. Frank Lipnizki

Call: Increasing capacity and skills of PhD students regarding industrially relevant neutron and synchrotron-based analytical methods – 2019
Project: Measurements of membrane fouling in lignocellulosic biorefineries by ptycho-tomography, 2019-03613

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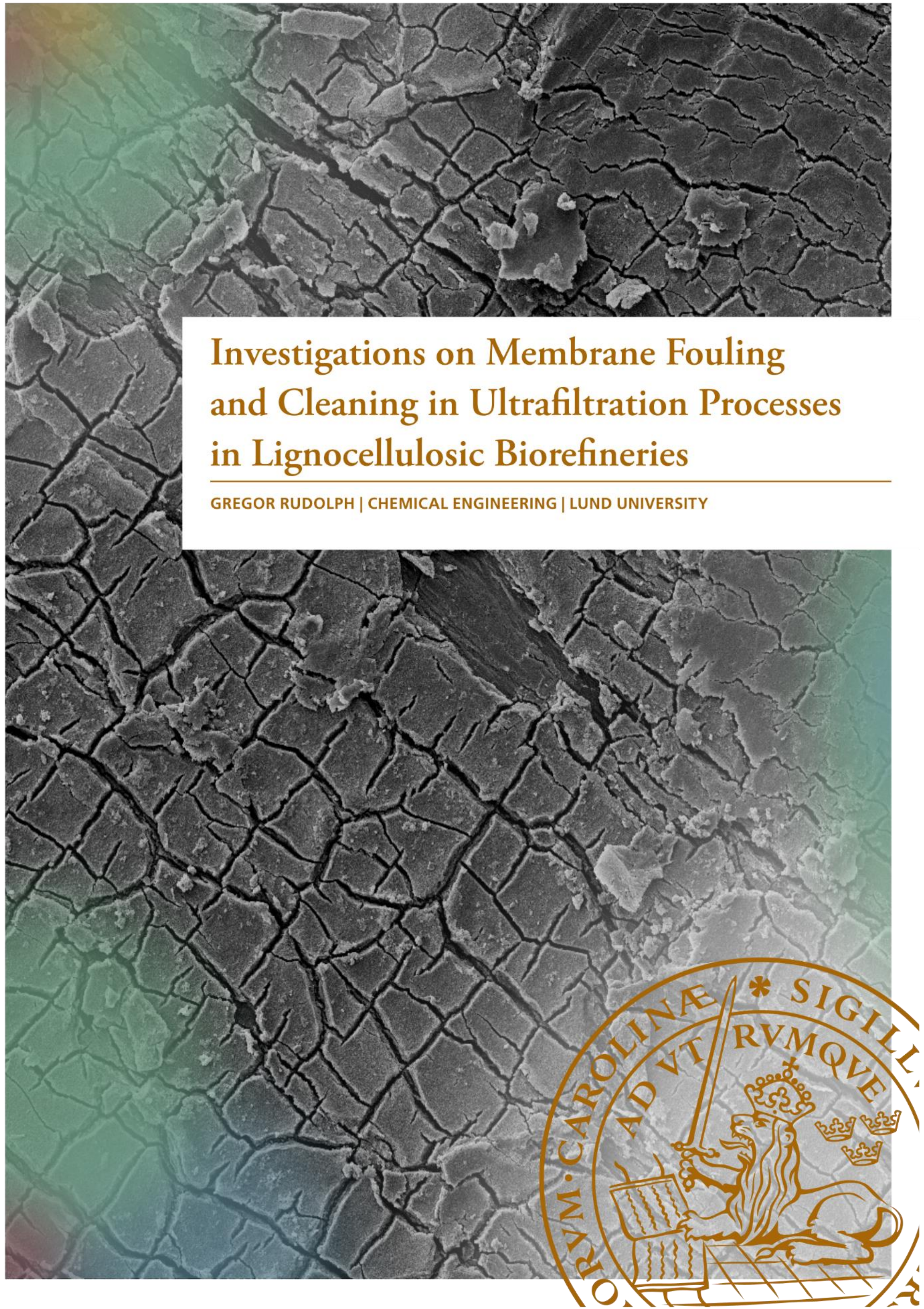

PhD defence tomorrow on membrane fouling and cleaning

When: 11 June, 9.15 am

Where: Livestreamed on YouTube

tinyurl.com/MembraneFouling