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Lithium Depth Profiling in Battery Anodes by Nuclear Reaction Analysis

Supporting Battery development and quality assurance using accelerators

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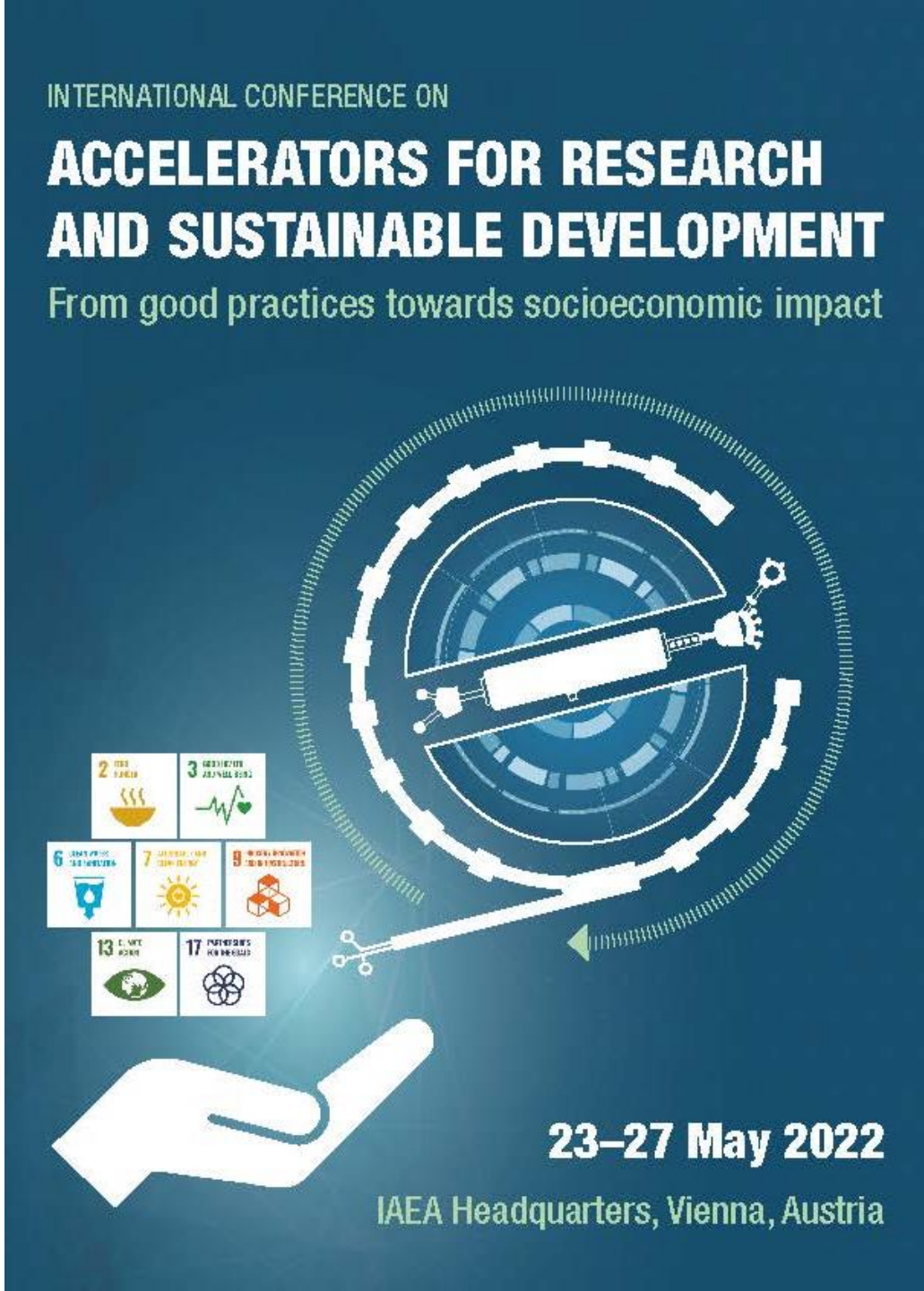
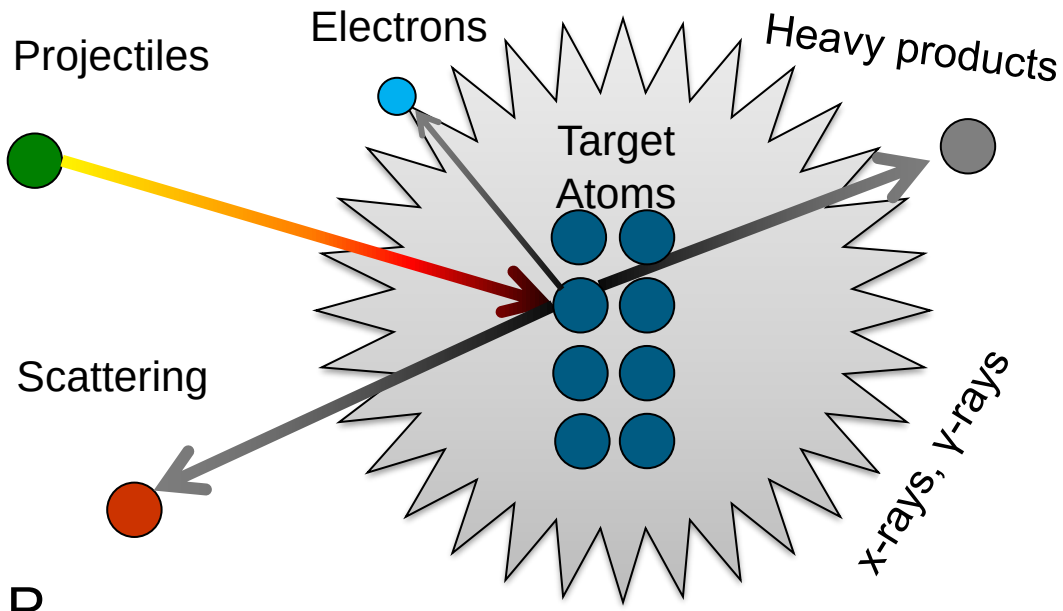


ION BEAM ANALYSIS

Relevant IBA methods for Li-Battery Research

- Rutherford Backscattering spectrometry RBS
  - ➢ Charged particle spectra with depth resolution
  - ➢ Quantification of heavier elements ≥C
- **Nuclear Reaction Analysis NRA** $^{7}Li(p,a)^{4}He$
  - ➢ Charged particle spectra with depth resolution
  - ➢ Quantification of light elements => Li
- Particle Induced X-ray Emission PIXE ≈ EDX
  - ➢ Good separation of medium elements Mn, Co, Fe, P…
- PIGE = Nuclear excitation PIXE
  - ➢ Quantification of light elements (Li, B, S, O, Al…)



Accelerator Technology,
Sören Möller, Springer, 2020

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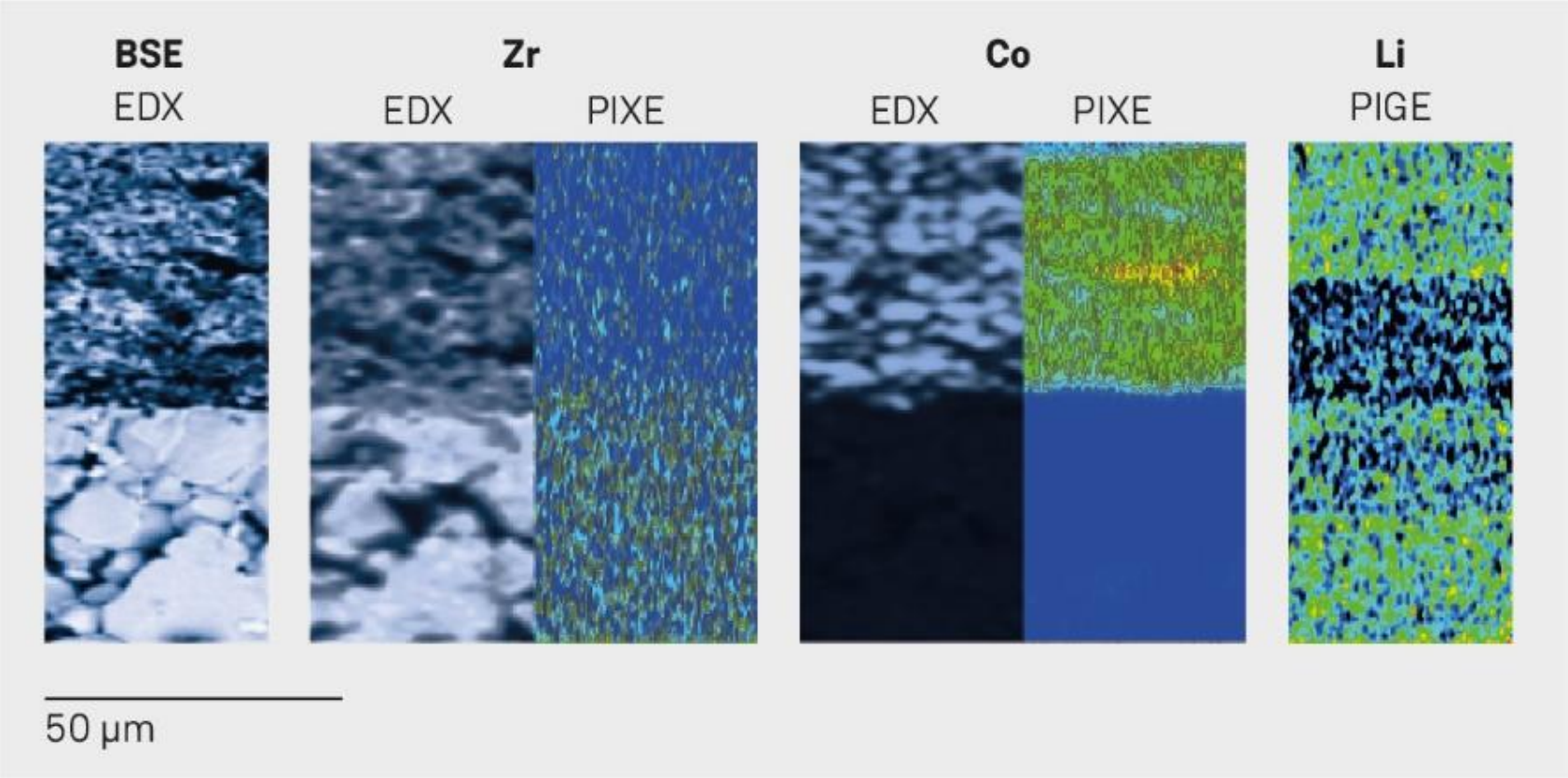
PROGRESS REPORT

Ion beam analysis for battery material analysis

Analysis of all-solid state batteries for determining SoH and SoC independently from electro-chemical analysis: https://doi.org/10.3390/batteries7020041

Latest paper for analysis of SiOx-Graphite anodes in coincells: https://doi.org/10.3390/batteries8020014



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SAMPLE PREPARATION

Si containing anodes

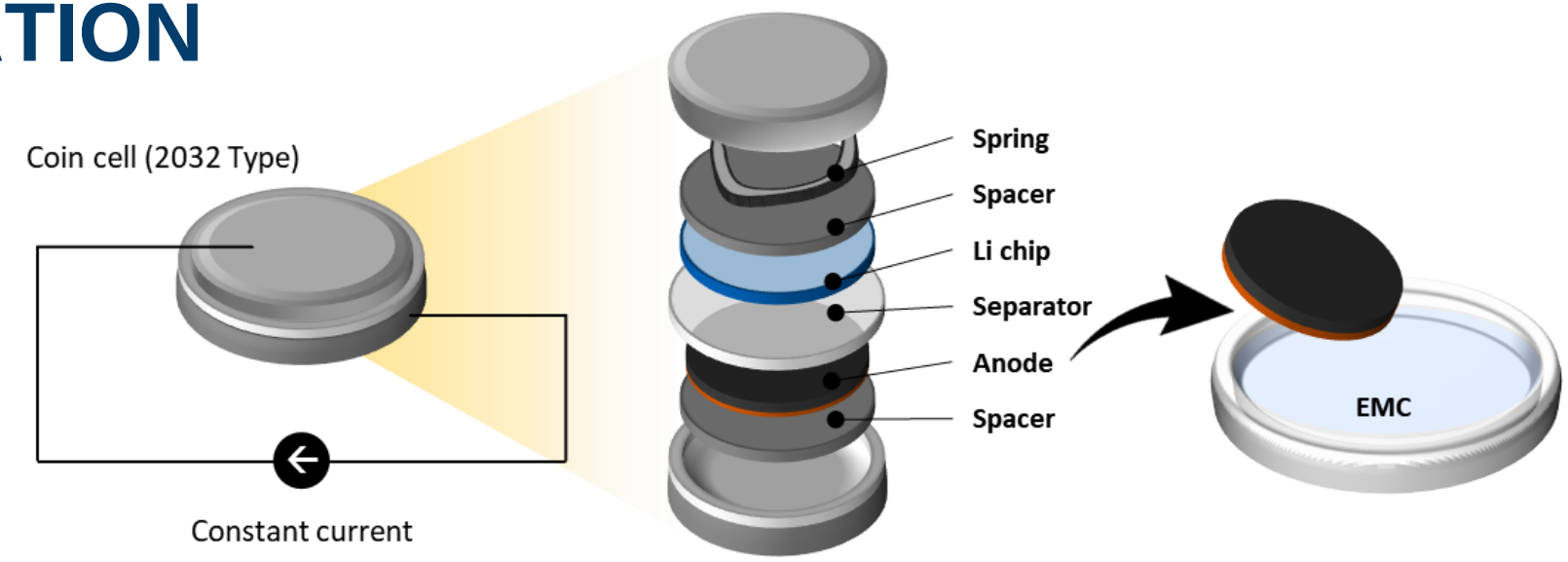
6 mixed Si-Graphite anodes prepared at 0-50% SoC

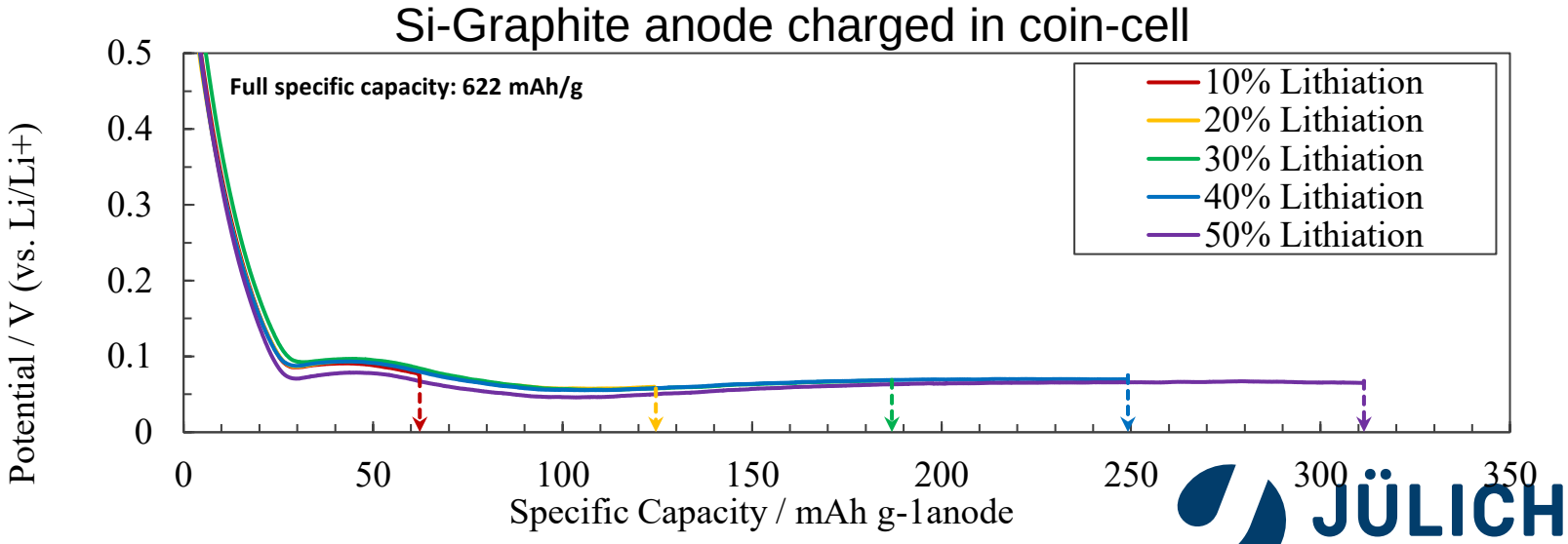
20 wt% SiOx resulting in 67% capacity increase vs. pure graphite

Anode thickness ~44 µm on Cu

Analysed ex-situ by IBA for

Full depth profiles for all elements





ION BEAM ANALYSIS DATA

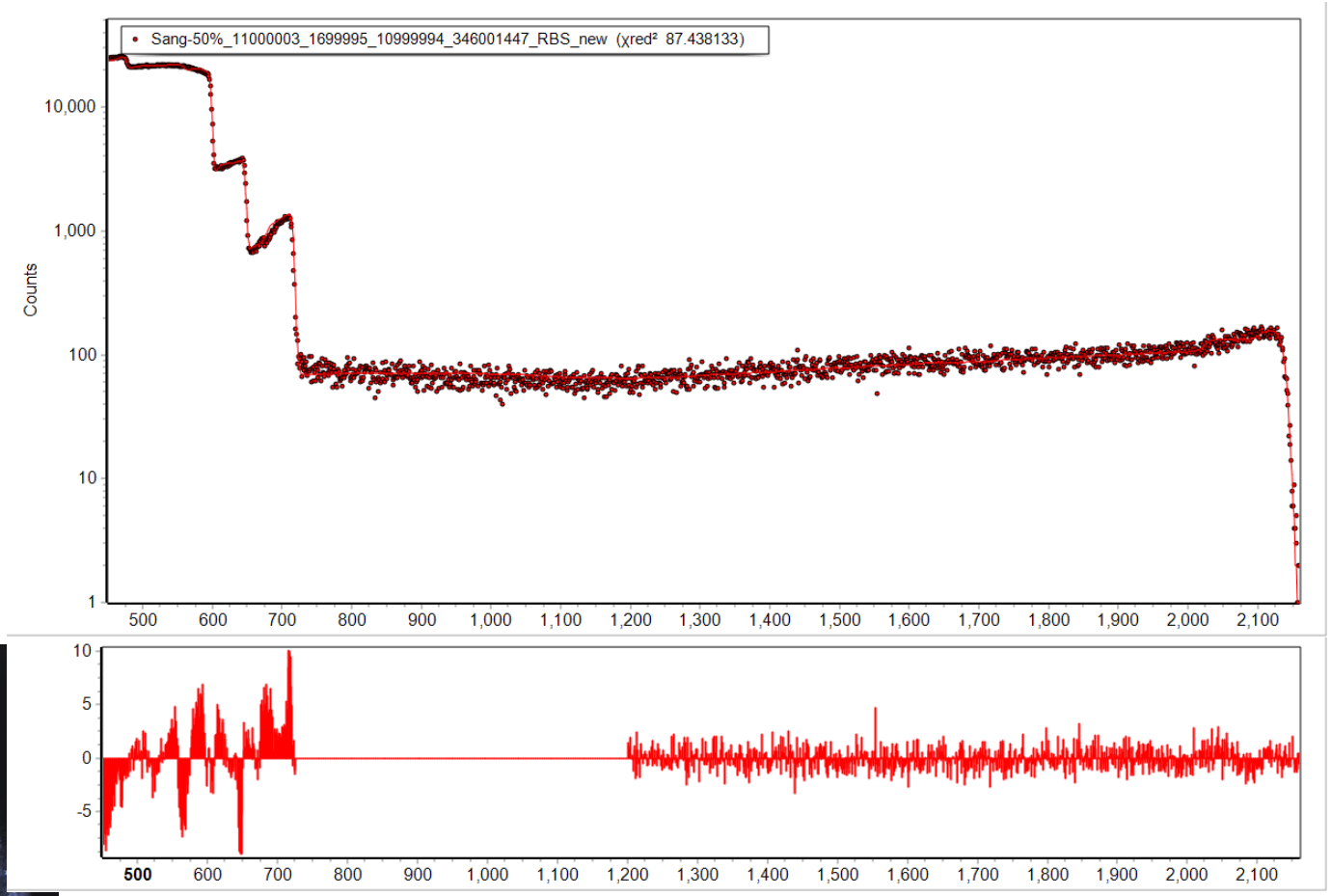
Setup details

2960 keV protons, ~5 nA current, beam spot diameter 2 mm

RBS + NRA with Multisimnra evaluation

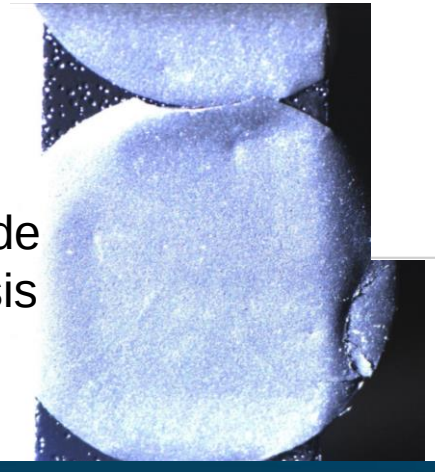
Example spectrum showing the 4 reaction edges for C, O, Si, Li

6 layers assumed for data fit



Example spectrum with fit and residuals

Photo of the anode disc in the analysis chamber



ION BEAM ANALYSIS DATA

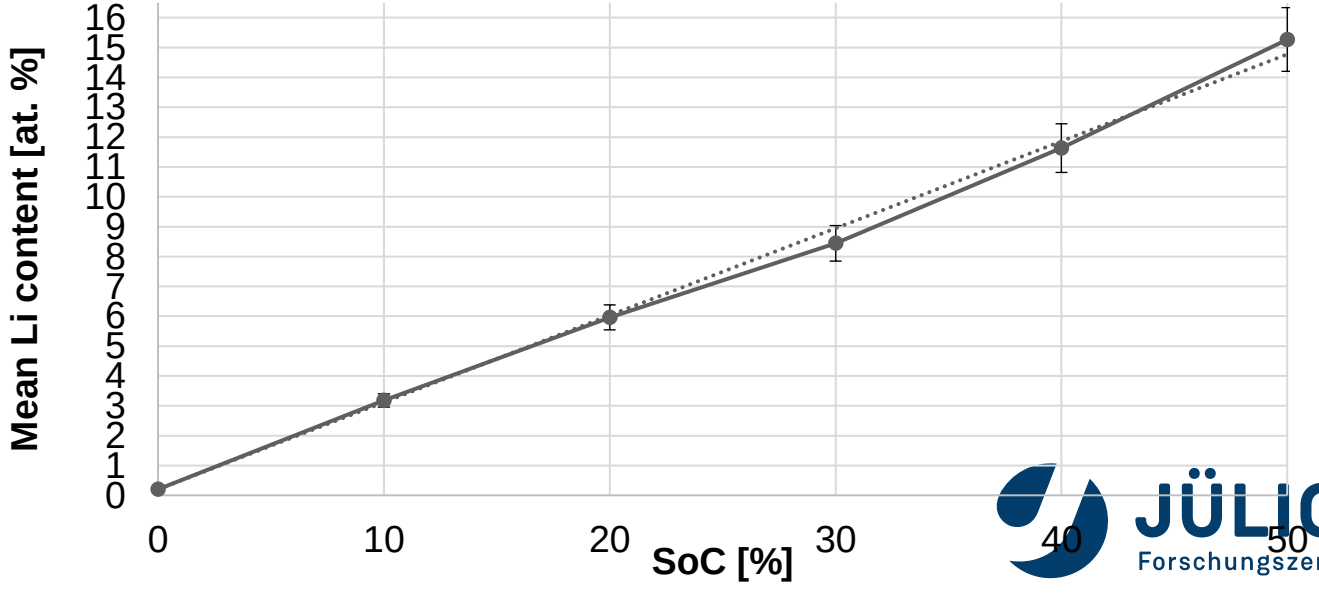
0D results

Perfect agreement between injected charge in mAh and detected Li atoms by IBA

Background Li from binder identified at SoC=0

Linear connection between SoC and integral Li content seen in IBA

Lithiation Degree/SoC (%)	Anode Capacity (mAh/cm²)	Lithiation charged (mAh)	Lithiation charged (10^{23} Li/m²)	Process Time (h)	IBA detected Li (10^{23} Li/m²)
0	3.78	0	**0.00**	0	**0.057**
10	3.78	0.38	**0.85**	0.5	**0.892**
20	3.78	0.76	**1.70**	1	**1.668**
30	3.78	1.13	**2.55**	1.5	**2.364**
40	3.78	1.51	**3.40**	2	**3.257**
50	3.78	1.89	**4.25**	2.5	**4.274**



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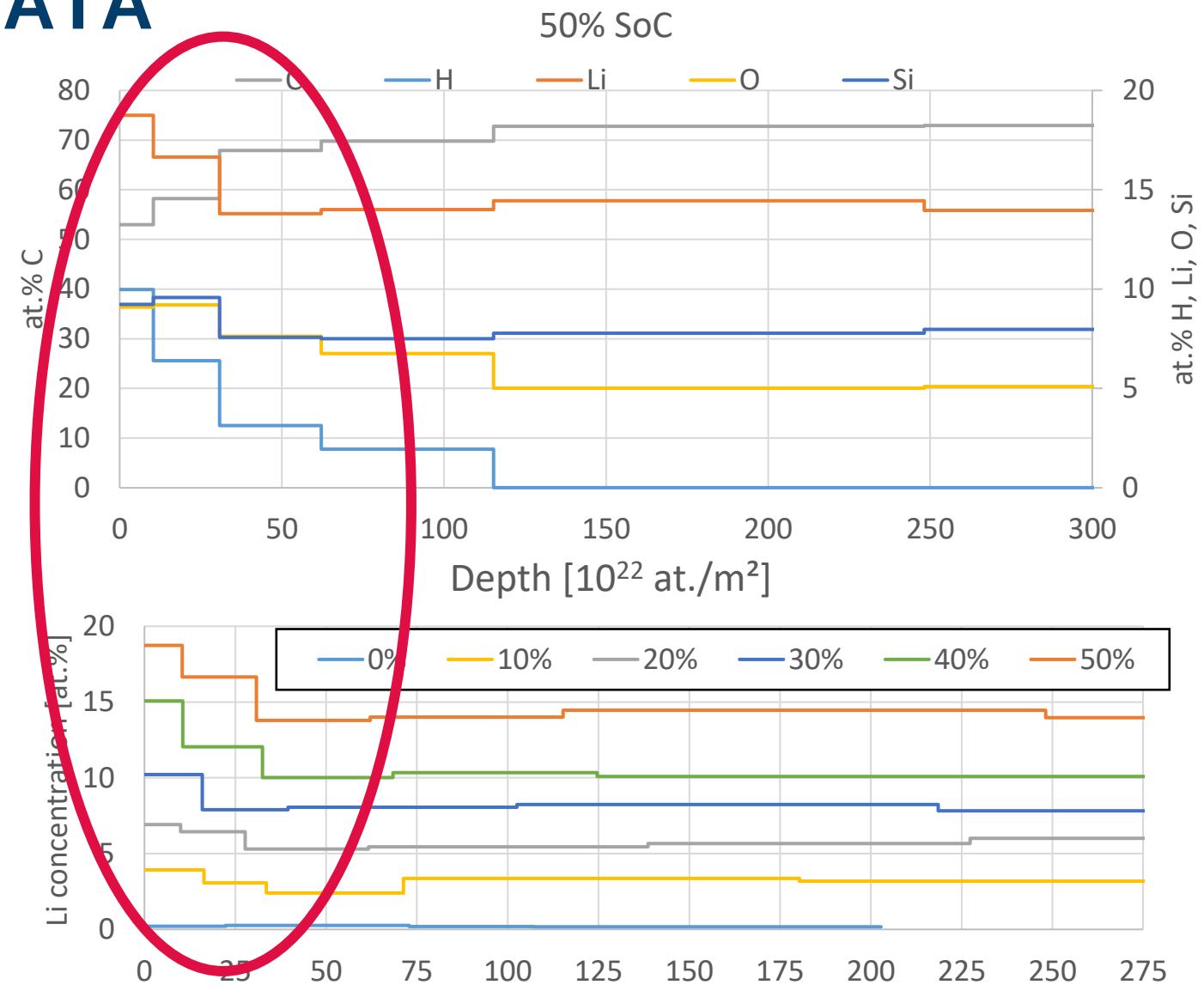
Depth profiles

Depth profiles of the 4 main elements in the anode show a 6 µm surface region and a constant bulk

Si, O, and Li enriched on the separator side of all anodes within first ~6 µm (=SiOx grain size)

H acts as a filling element in the surface layer (only invisible element for analysis)



ION BEAM ANALYSIS DATA

SiO_x distribution in graphite anodes

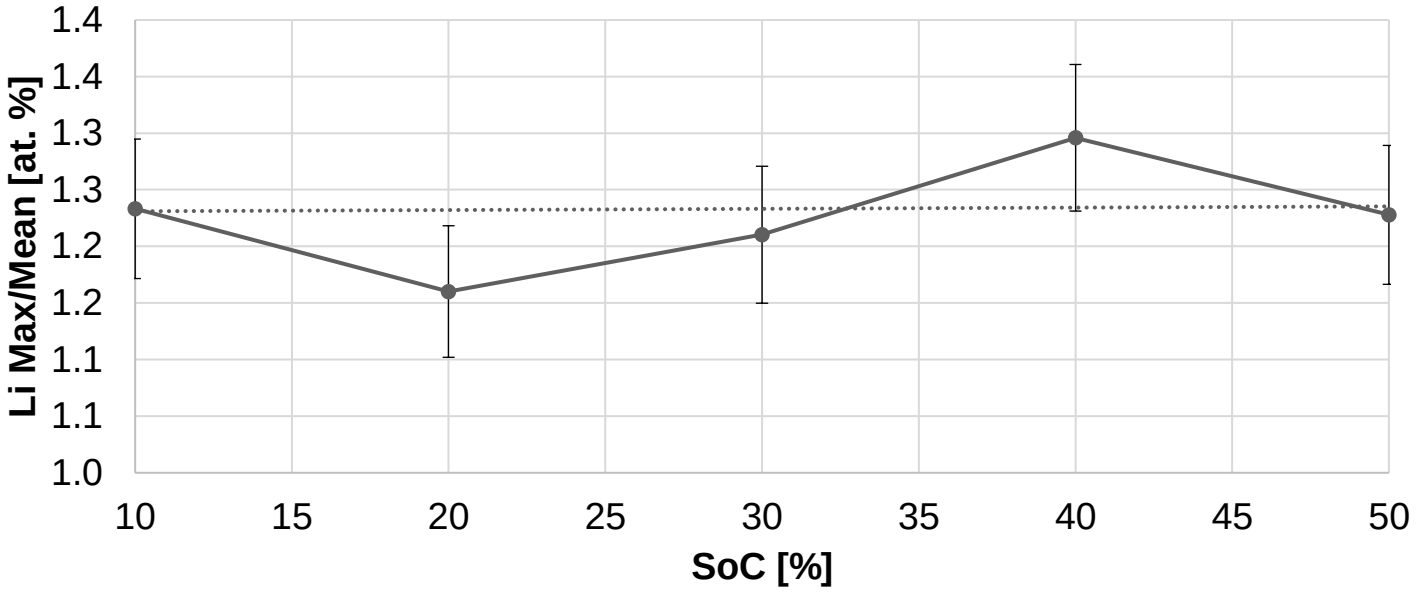
Enrichment of Si, O, Li at the separator side always a factor 1.23 compared to the bulk

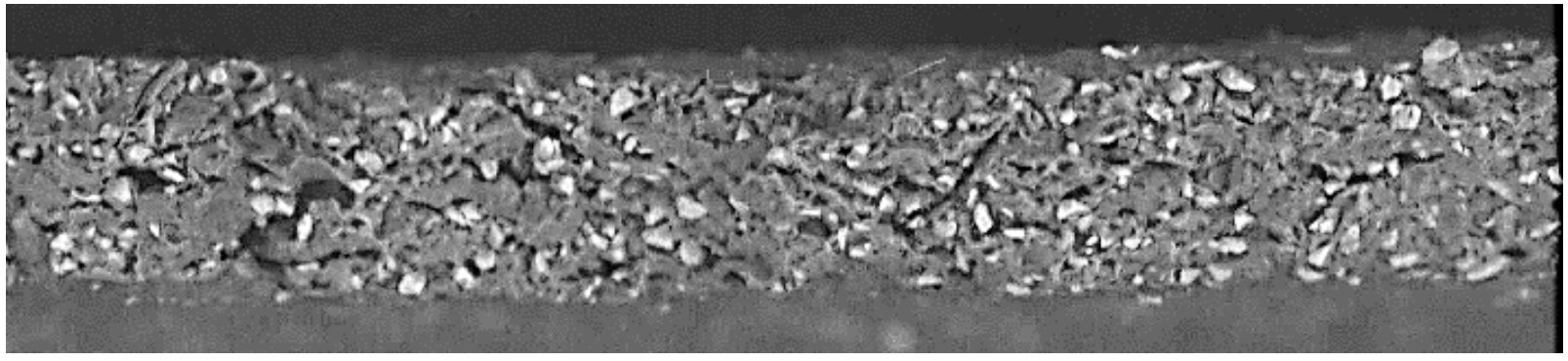
Si, O depth profiles also show separator side peak, but might be Al from separator coating instead

SEM cross-cut analysis resolution insufficient to exclude SiOx enrichment, but found Al

Li preferably binds to Si not C

Maximum Li/Si=1.04±0.05 derived for given SiOx





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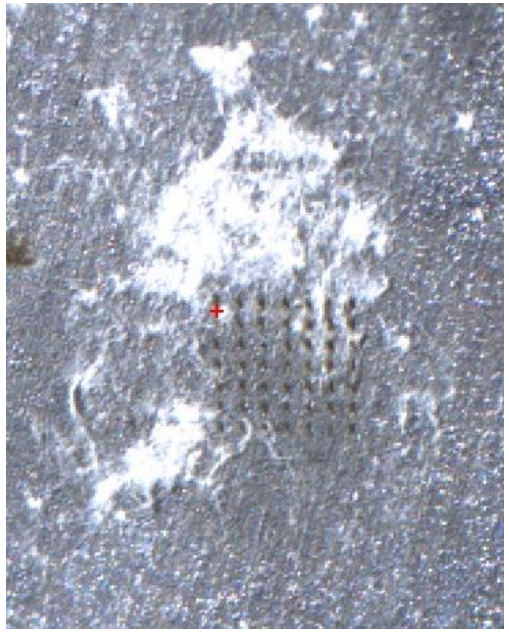
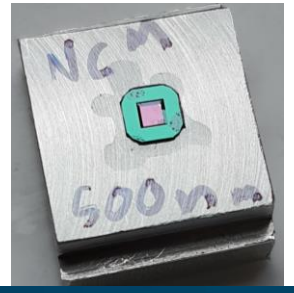
SUMMARY

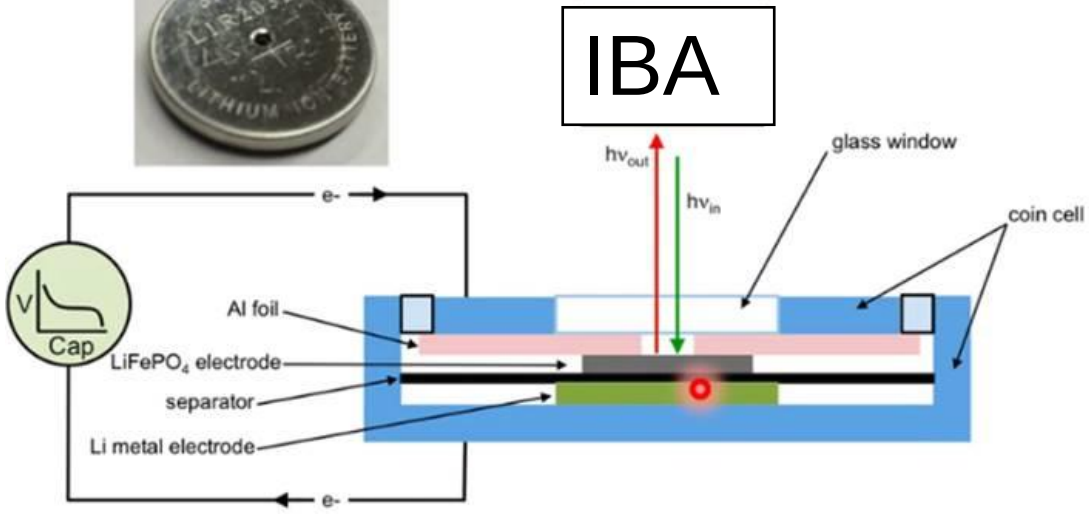
- Anodes successfully measured ex-situ
- Good agreement of Li amount between IBA and EC
- Depth profiles with ~ µm resolution of all constituents
- Revealed separator side Li enrichment in first 6 µm with factor 1.23
- Derived maximum loading of 1:1 Li/Si for SiO_x



Outlook:

- IBA Startup (see exhibition)
- Comparing IBA to other methods
- In-situ coin cell analysis using IBA (picture)





Thank you

Thanks to Christian Scholtysik for operating the Tandem accelerator