ENABLING RESEARCH PROJECT ENR-MAT.01.VR

Electronic interactions of slow ions and their influence on defect formation & sputter yields for plasma facing components

E. Pitthan[1], M. V. Moro[1], J. Shams-Latifi[1], P. M. Wolf[1], B. Bruckner[1], T. Tran[1], D. Moldarev[1], P. Ström[1], D. Primetzhofer[1], P. Petersson[2], M. Rubel[2], C. Cupak[3], M. Fellinger[3], F. Aumayr[3], L. Caveglia Curtil[4], E. Ponomareva[4], T. Malykhina[4], A. Aro[4], A. Sand[4]

[1]Department of Physics and Astronomy, Uppsala University, 751 20 Uppsala, Sweden
[2]Department of Fusion Plasma Physics, KTH Royal Institute of Technology, 100 44 Stockholm, Sweden
[3]TU Wien, Institute of Applied Physics, Fusion@ÖAW, 1040 Vienna, Austria
[4]Department of Applied Physics, Aalto University, Aalto, Espoo FI-00076, Finland

Monitoring of 2022 Enabling Research activities, 07th of February 2023

This work has been carried out within the framework of the EUROfusion Consortium, funded by the European Union via the Euratom Research and Training Programme (Grant Agreement No 101052200 — EUROfusion). Views and opinions expressed are however those of the author(s) only and do not necessarily reflect those of the European Union or the European Commission. Neither the European Union nor the European Commission can be held responsible for them.

Outline

☐ Aim;

☐ Working-packages & team;

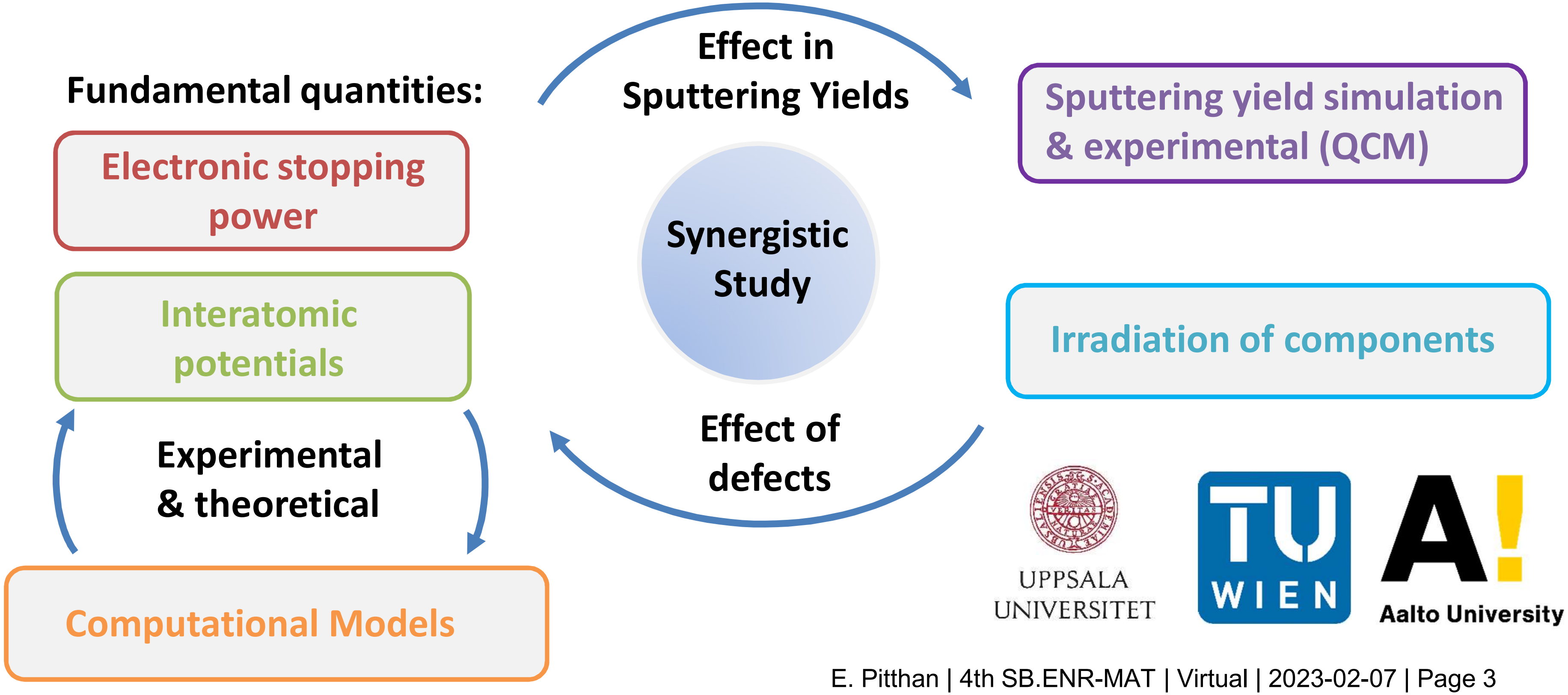
☐ Main results obtained & work in progress;

☐ Achievement of Scientific Deliverables foreseen for 2022;

☐ Activities foreseen for 2023.

Aim

To investigate underlying **quantities fundamental** for **sputtering** and **defect formation** from plasma-wall interaction: important input variables in modelling of **erosion** and **implantation** in plasma facing components.



Fundamental quantities:

Electronic stopping power

Interatomic potentials

Experimental & theoretical

Computational Models

Effect in Sputtering Yields

Synergistic Study

Effect of defects

Sputtering yield simulation & experimental (QCM)

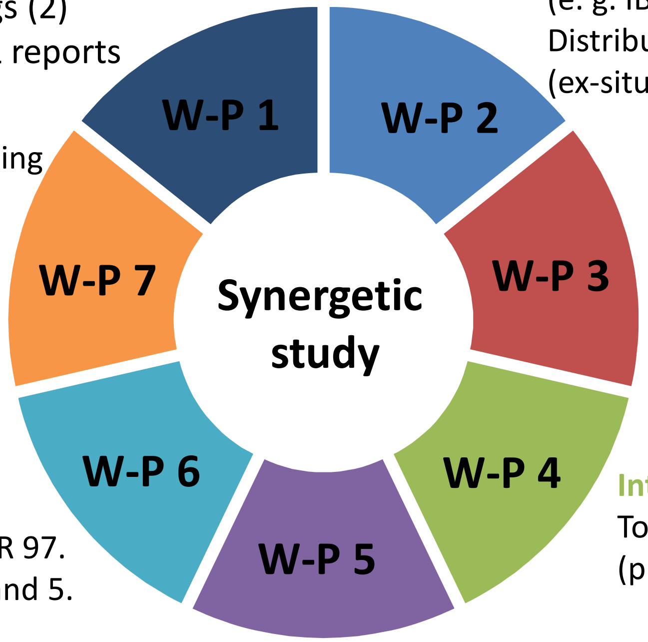
Irradiation of components

UPPSALA UNIVERSITET

TU WIEN

Aalto University

Working-packages



General management
Kick-off meeting
Task coordination
Annual meetings (2)
Final meeting & reports

Sample preparation
Batches of Fe, W, and EUROFER97.
Chemical and morphological characterization (e. g. IBA; AFM; XRD).
Distribution and routinely quality control (ex-situ and in-situ).

Electronic energy loss measurements
Experimental stopping cross-section of pristine Fe, W, and EUROFER97.
Energy range: sub-keV to MeV.
Compare experimental results between pristine & damaged.

Interatomic potential measurements
ToF-LEIS & ToF-MEIS angular scans measurements (pristine and damaged samples).

Sputtering yield & QCM
QCM set-up at UU. Sputtering yield and angular distribution of pristine Fe, W, and EUROFER 97 samples. BCA-based simulations (SDTrimSP).

Ion-irradiation experiments
Ex-situ and In-situ ion irradiation experiments on Fe, W, and EUROFER 97.
Samples to be reused in W-Ps 3,4, and 5.

Computational modelling
TDDFT simulations of electronic stopping power of W, Fe, and EUROFER-like (with/without defects).
MDRANGE: ion implantation & Sputter yields (Input: electronic stopping)

Team

VR	ÖAW	VTT
Eduardo Pitthan (PI)	Christian Cupak	Andrea Sand
Jila Shams-Latifi	Martina Fellinger	Ludovico Caveglia Curtil
Petter Ström	Alexander Redl	Tetiana Malykhina
Per Petersson	Friedrich Aumayr	Akseli Aro

Start: May of 2021.

VR main tasks: Sample preparation and characterization, electronic loss measurements, interatomic potential measurements, and ion irradiation experiments.

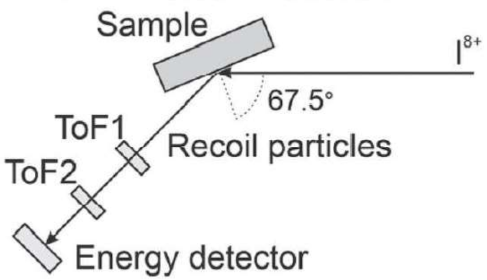
ÖAW main tasks: Sputtering yield measurements, and BCA-based simulations.

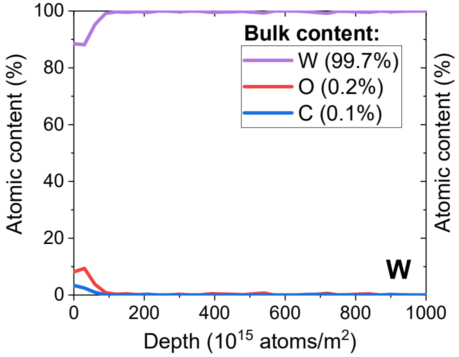
VTT main tasks: Computational modelling (simulations of electronic stopping power & ion implantation).

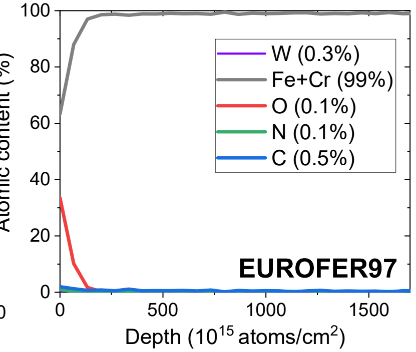
W-P 2: Sample preparation

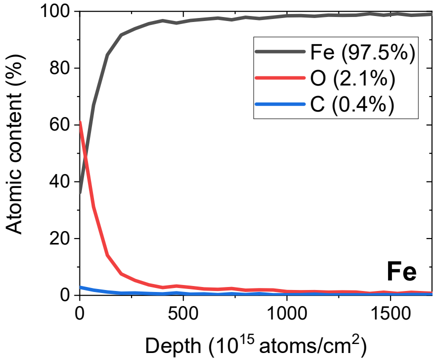
Characterization of the chemical composition of the pristine samples (Fe, W, EUROFER) by combined ion beam based techniques (UU), as a protocol for the standard quality control.

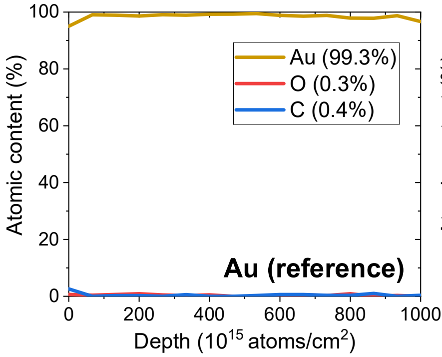
→Atomic concentration depth profiles by ToF-ERDA:

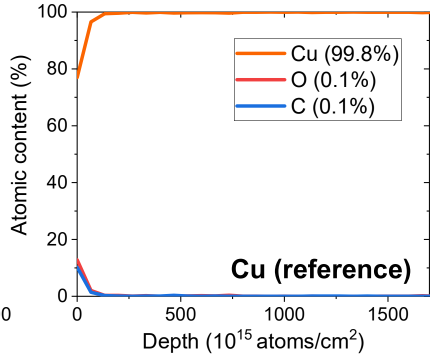












→Low impurity concentrations in bulk.

Evaluation window 500-1000×10^{15} atoms/cm^2

Final stopping data corrected by Bragg's rule:

$$\epsilon^{A_m B_n \cdots} = m\epsilon^A + n\epsilon^B + \cdots$$

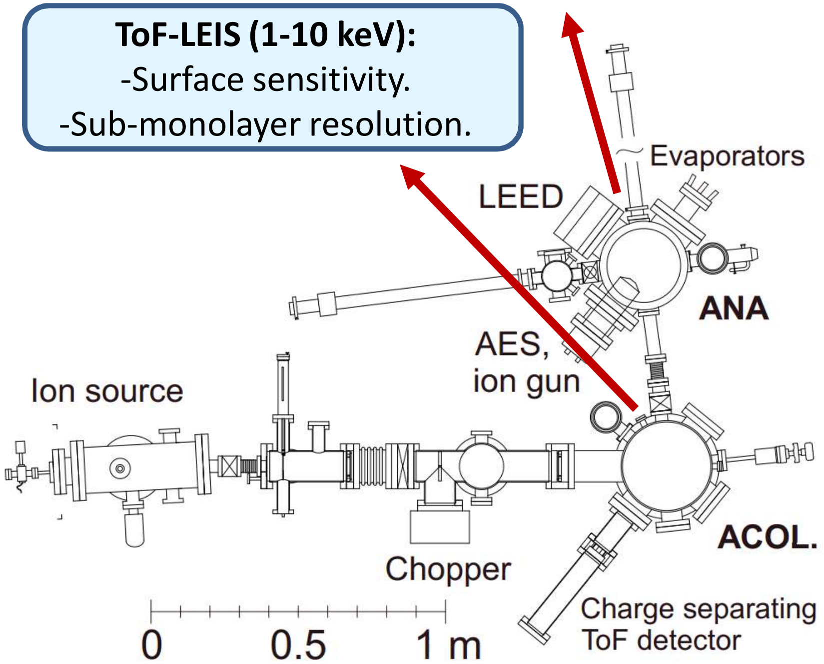
W-P 3: Electronic energy loss measurements

Experimental procedure for low energy regime

ACOLISSA experimental set-up:

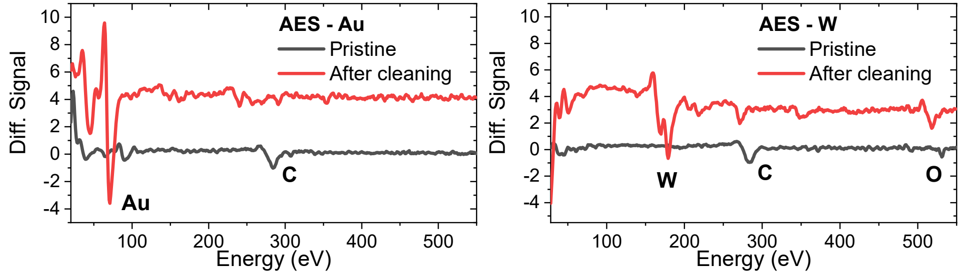
Analytical Chamber: Sputtering cleaning, annealing, AES, e$^-$beam evaporation, and LEED.

ToF-LEIS (1-10 keV):
-Surface sensitivity.
-Sub-monolayer resolution.

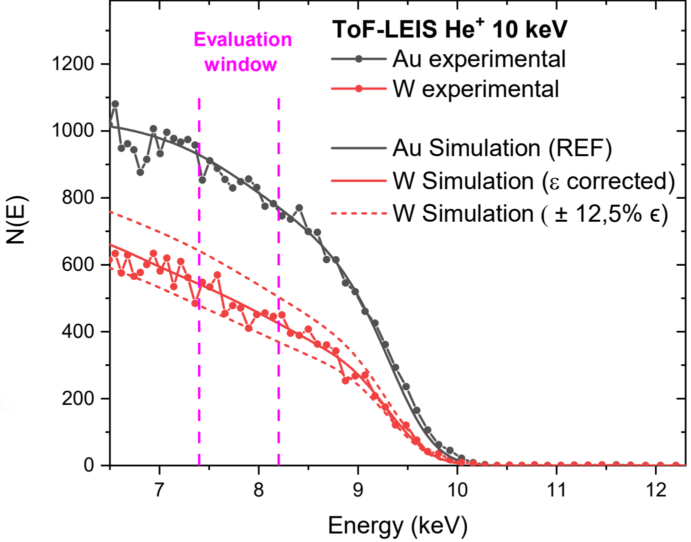


P. Ström and D. Primetzhofer 2022 JINST 17 P04011

Sample cleaning: *cycles of Ar$^+$ sputtering 3 keV 30˚.*



SCS relative measurements in comparison with MC simulations:



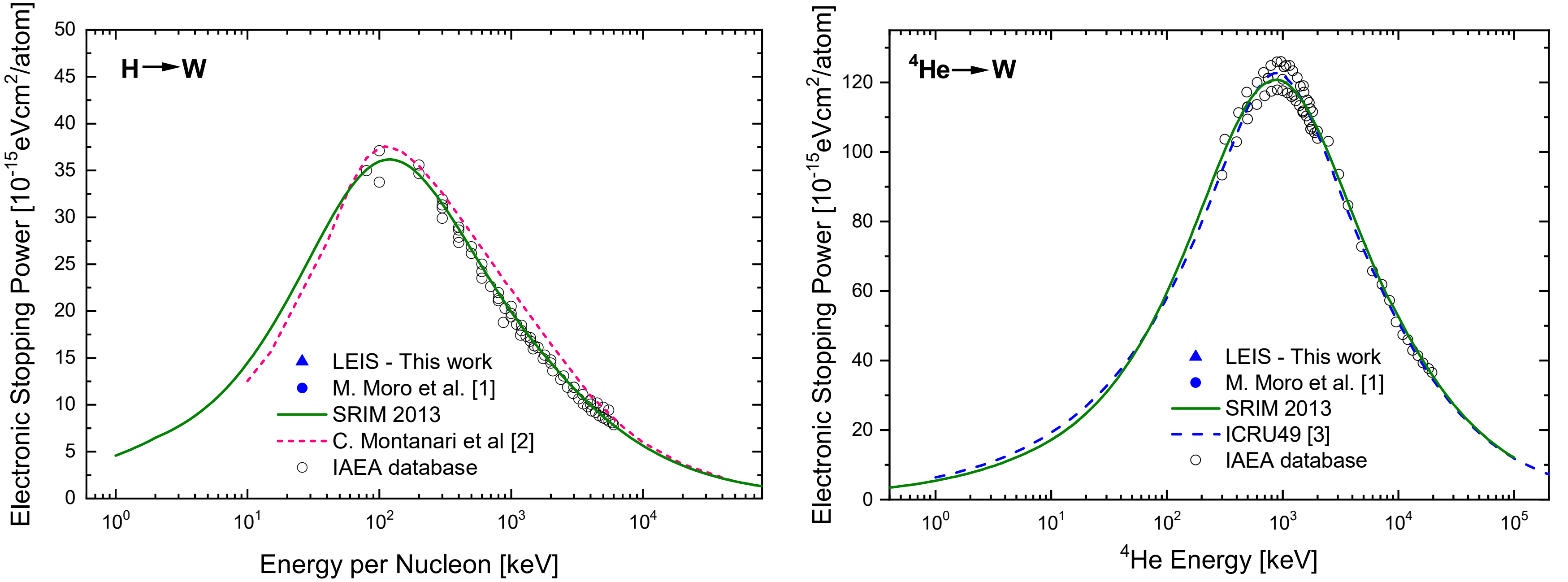
→Cu/Au as reference under same experimental condition.

→SCS extraction from height ratio.

→ Similar approach for MEIS and MeV energies our laboratory.

W-P 3: Electronic energy loss measurements

Experimental stopping cross-section of pristine Fe, W, and EUROFER97



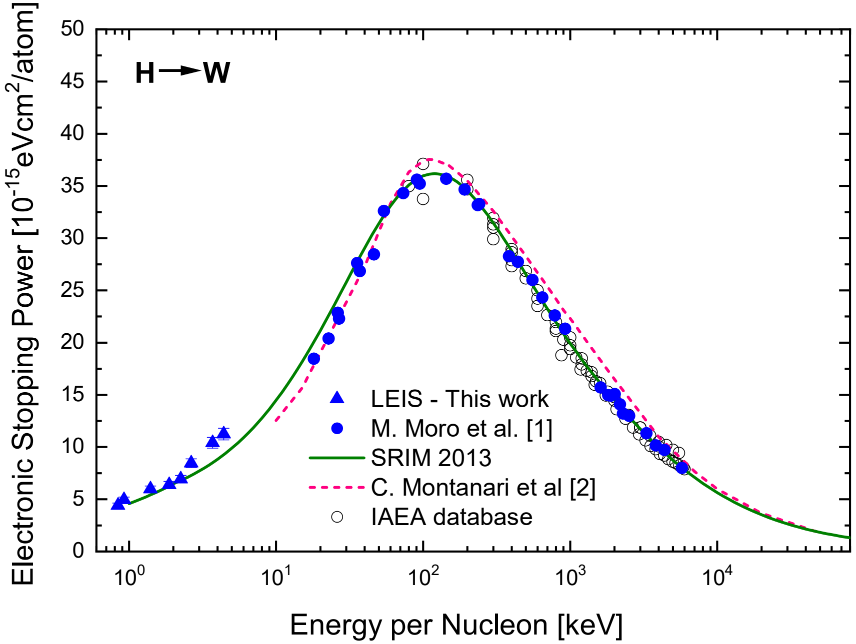
[1] M. V. Moro et al., Nucl. Instrum. Meth B **498** (2021).

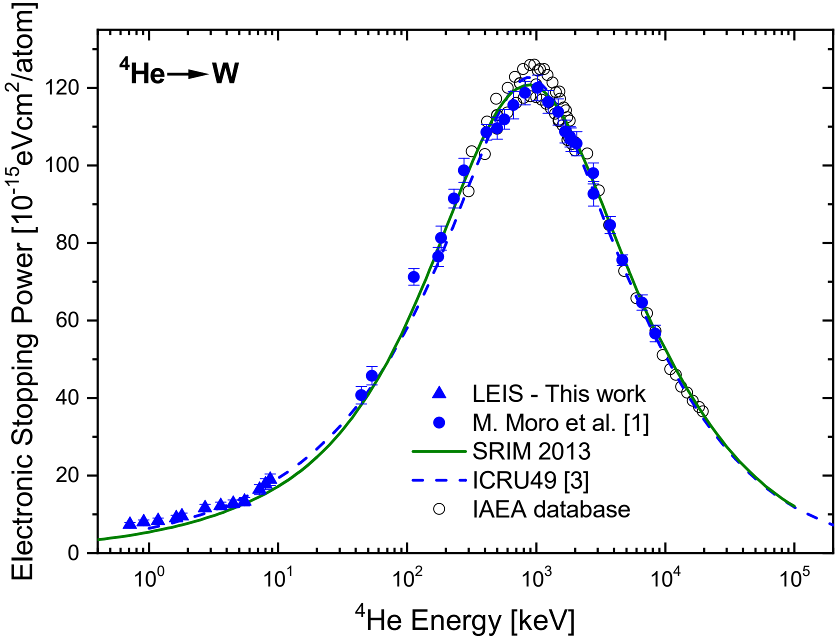
[2] C. C. Montanari et al., Phys. Rev. A **80**, 012901 (2009).

[3] M. J. Berger, et al., Report 49, Oxford Academic (1993).

W-P 3: Electronic energy loss measurements

Experimental stopping cross-section of pristine Fe, W, and EUROFER97





MEIS and MeV Range [1]: Good agreement with experimental and SRIM 2013 (up to 3.5% for protons and 4.0% for He).

LEIS: Discrepancies from SRIM-2013 up to 20% for protons and 60% for He (good agreement with ICRU49).

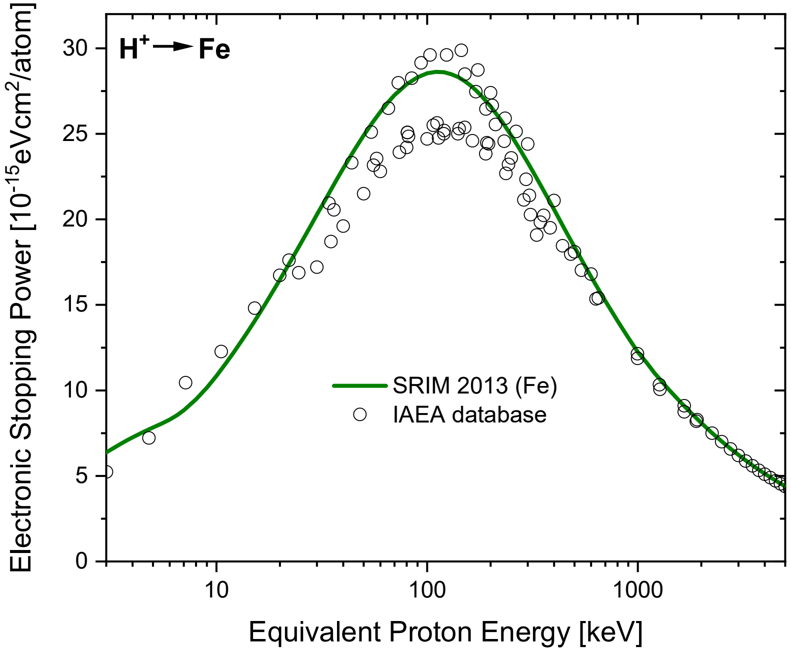
[1] M. V. Moro et al., Nucl. Instrum. Meth B **498** (2021).

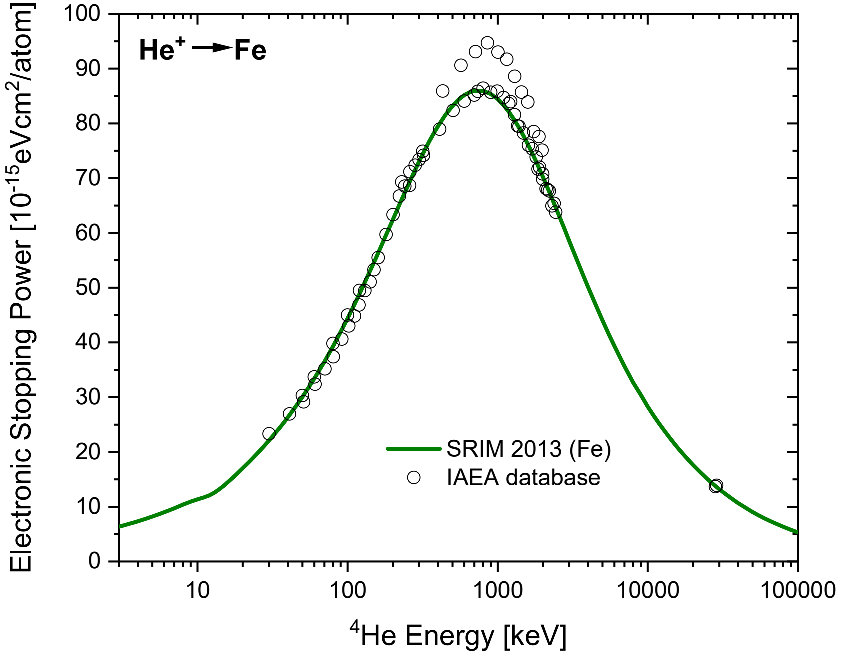
[2] C. C. Montanari et al., Phys. Rev. A **80**, 012901 (2009).

[3] M. J. Berger, et al., Report 49, Oxford Academic (1993).

W-P 3: Electronic energy loss measurements

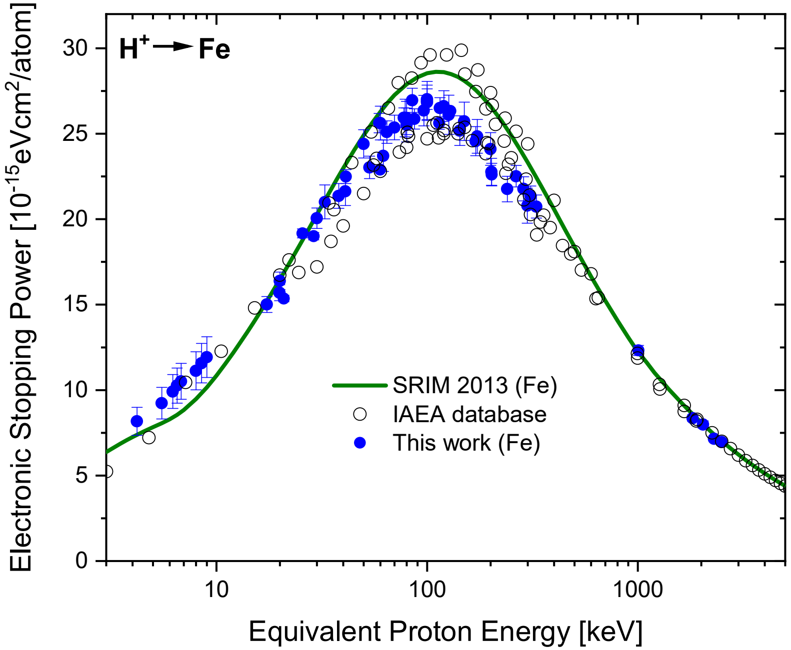
Experimental stopping cross-section of pristine Fe, W, and EUROFER97





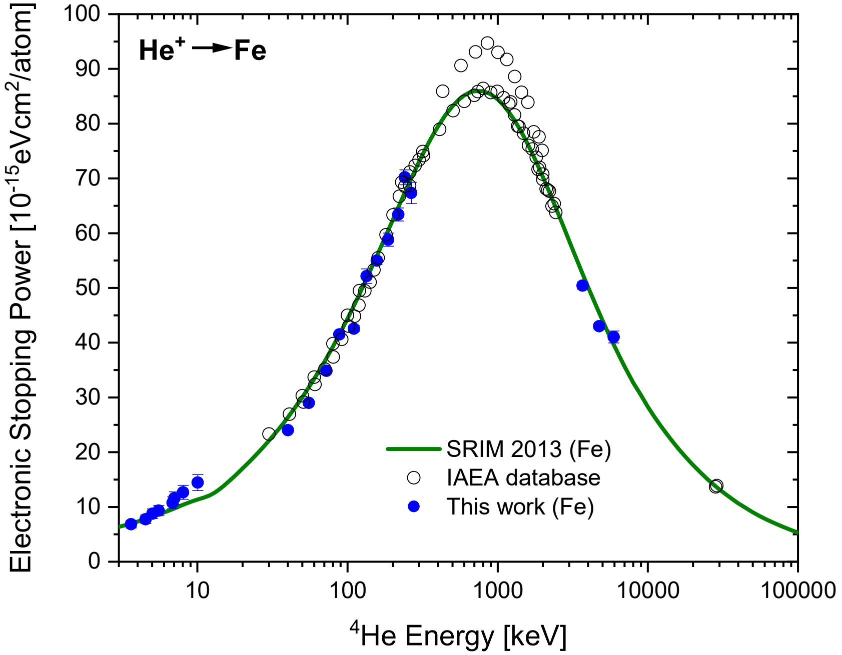
W-P 3: Electronic energy loss measurements

Experimental stopping cross-section of pristine Fe, W, and EUROFER97



H$^+$ → Fe

→ Good agreement at MeV range.
→ Large discrepancy with SRIM around maximum (up to 11%).
→ Large discrepancy with SRIM at low energy range (up to 20%).

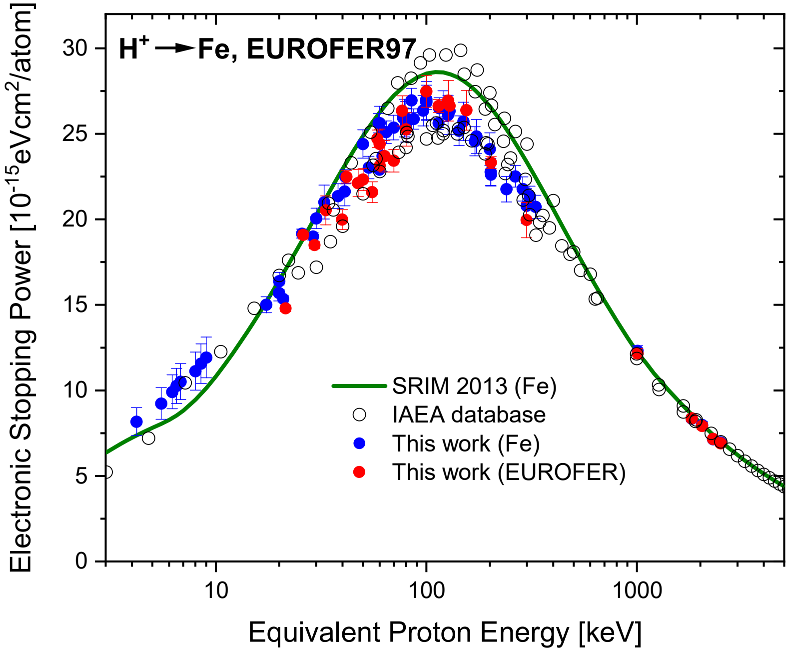


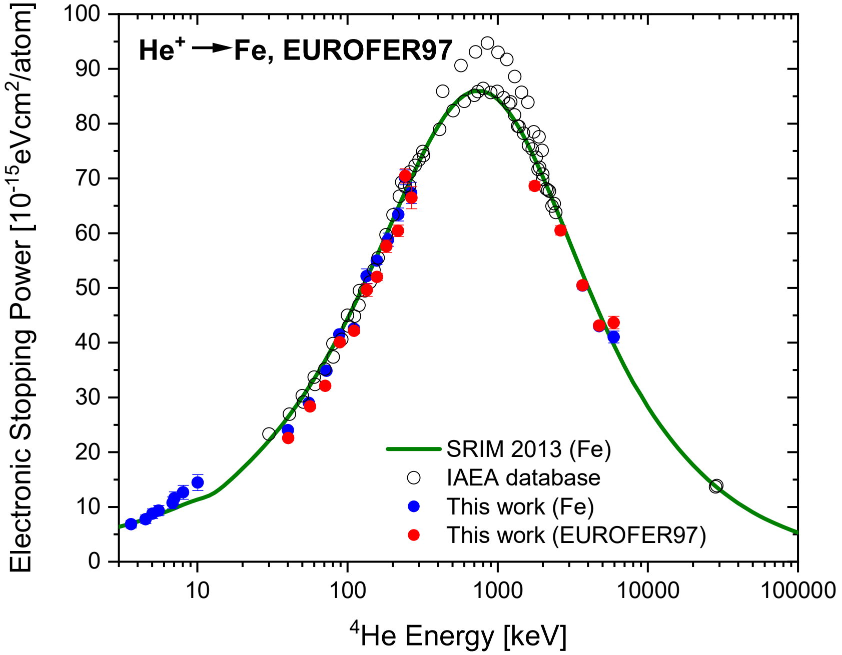
He$^+$ → Fe

→ Good agreement at keV/MeV range within 6%.
→ LEIS: discrepancy up to 26%.

W-P 3: Electronic energy loss measurements

Experimental stopping cross-section of pristine Fe, W, and EUROFER97





H^+, He^+ → EUROFER97

→ EUROFER97: similar SCS to Fe.

→ No clear deviation from Bragg's rule

In progress (2023):

→ Intermediate energies (400 to 1.8 MeV) [HZDR].

→ W and EUROFER97 SCS using thin films (absolute approach).

W-P 4: Interatomic potential measurements

Data and analysis: P. W. Wolf, and E. Pitthan

Crystal samples received (March 2022):
W(110), Fe(100), Cr(100)

- **In the LEIS regime (1-10 keV):**
- **Scattering potential (Thomas-Fermi-Molierè):**

$$V(r) = \frac{Z_1 Z_2 e^2}{r} . \underset{\uparrow}{\Phi}(\frac{r}{\underset{\uparrow}{a}})$$

Screening function

Screening lenght

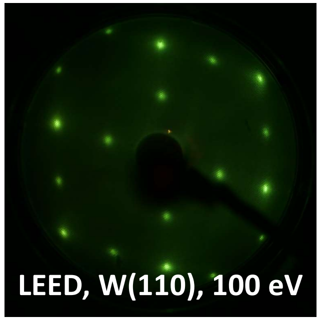
$$a = \underset{\uparrow}{c_a} a_f$$

Screening correction factor (empirical)

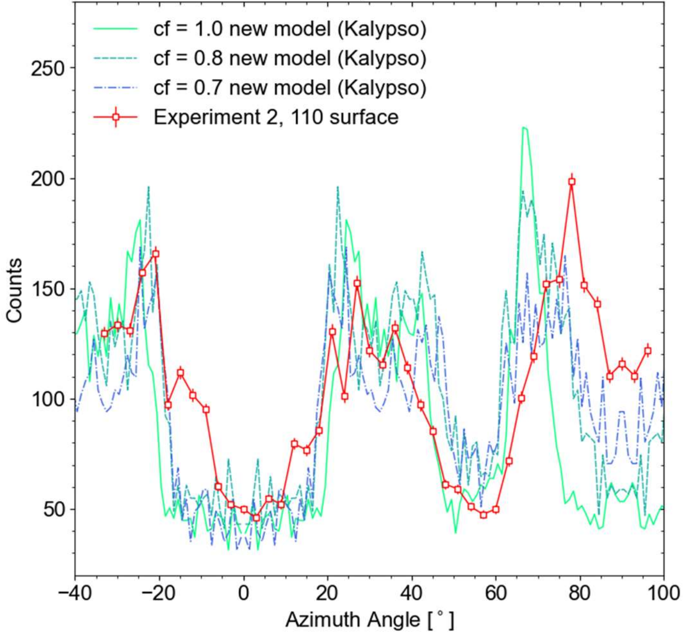
For a given θ:
$c_a < 1$ → smaller scattering cross-section.

The plan:
Measuring the interatomic potential of W, Cr, and Fe by comparing angular LEIS scans with KALYPSO simulations.

Experiment:
- W(110) sample
- Multiple cycles of sputter cleaning and annealing
- Azimuth scan from 0° to 132° using 3 keV He⁺


LEED, W(110), 100 eV

ToF-LEIS, 3 keV He^+

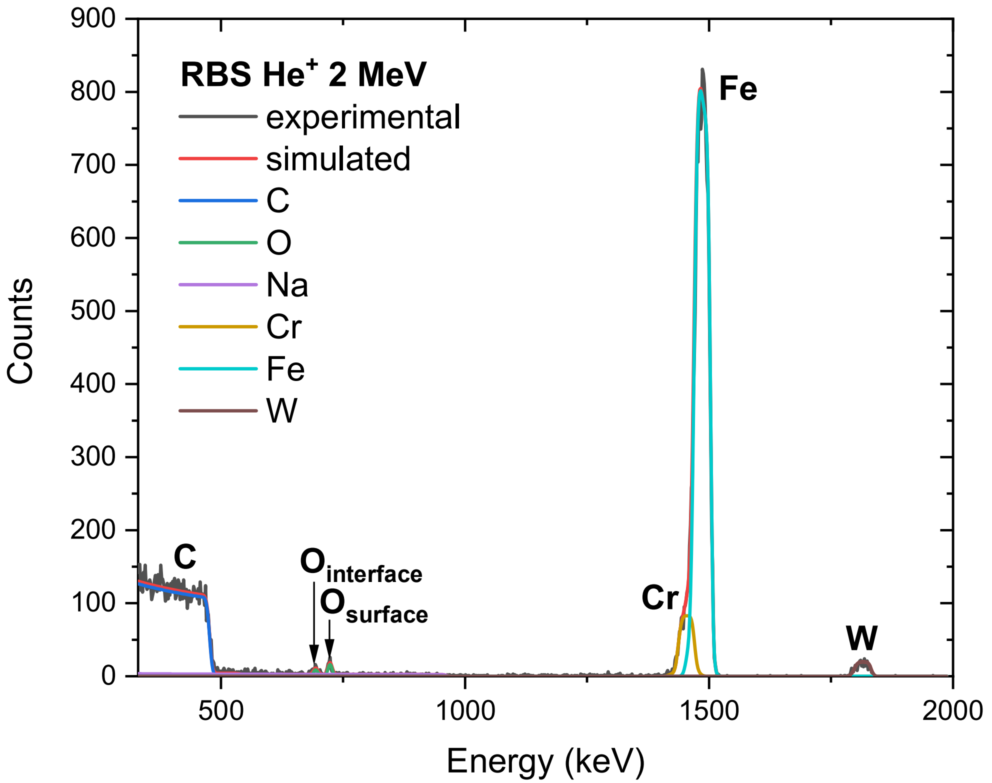


Experiments and simulations are in progress.
Simulations:
- Using KALYPSO, a software for molecular dynamics simulation of atomic collisions in solids.
- Using Thomas-Fermi-Moliere potential under different corrections.

W-P 5: Sputtering yields and BCA simulation

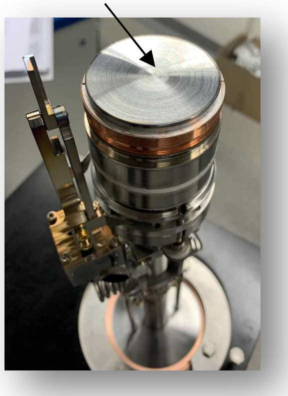
Formation and characterization of thin films from EUROFER97 target

In collaboration with P. Petersson and M. Rubel [1]



[1] KTH Royal Institute of Technology, Stockholm, Sweden

Sputtering target (2 inches):


EUROFER97

Sputtering conditions:

P_{base} = 7.1×10^{-8} mbar

P_{Ar} = 5.61×10^{-3} mbar

f_{Ar} = 10 sccm

P = 25 W

$Rate_{QCM}$ = 4.91 nm/min

From SIMNRA: Similar composition to bulk.

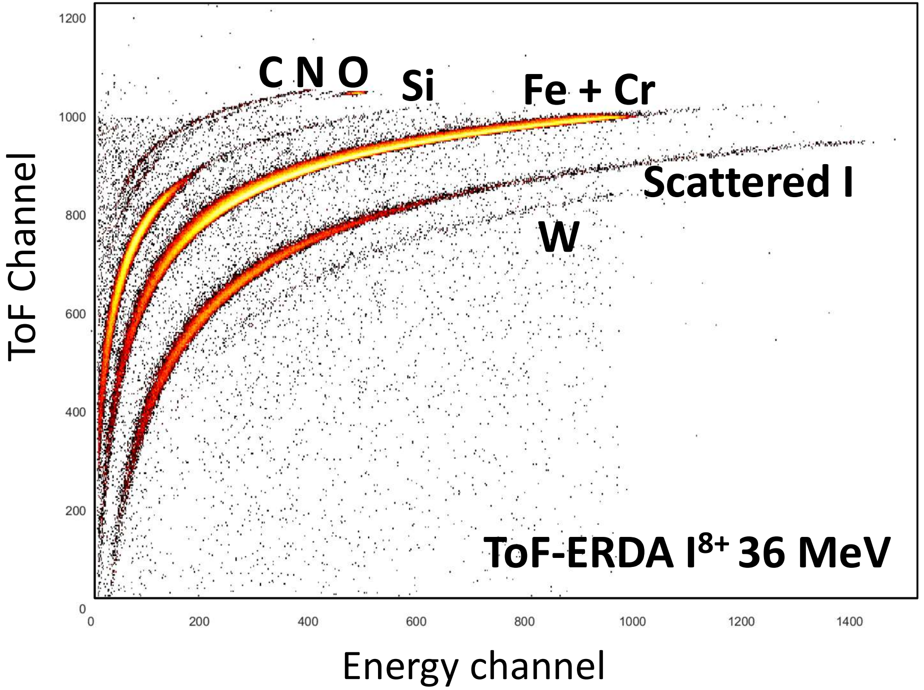
	At. content (%)	
	Sputtering	Bulk (nominal)
Fe	88.7	88.9
Cr	11.0	9.5
W	0.3	0.3

W-P 5: Sputtering yields and BCA simulation

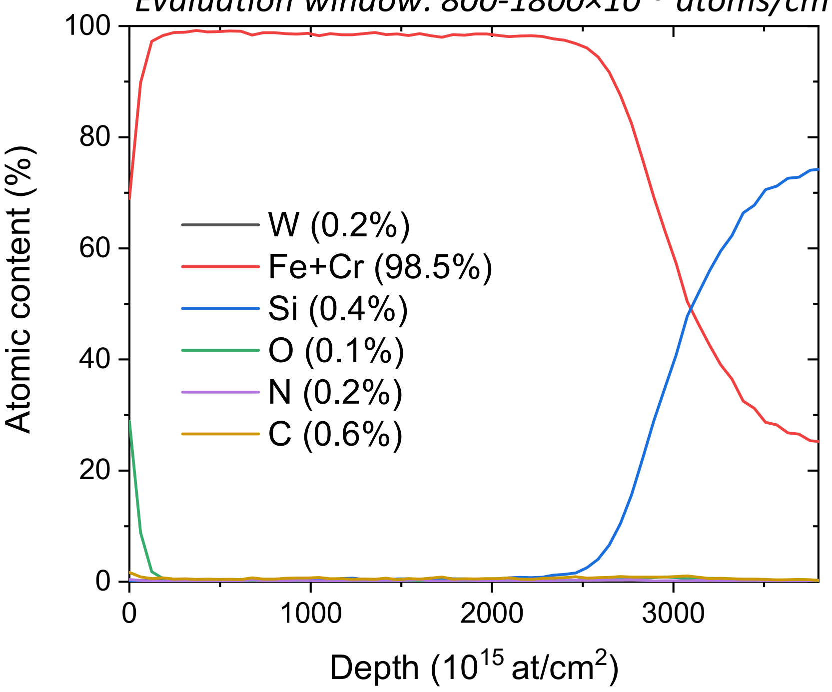
Formation and characterization of thin films from EUROFER97 target

In collaboration with P. Petersson and M. Rubel [1]

EUROFER97 Sputtering (60 min) on Si substrate



Evaluation window: 800-1800×10^{15} atoms/cm^2

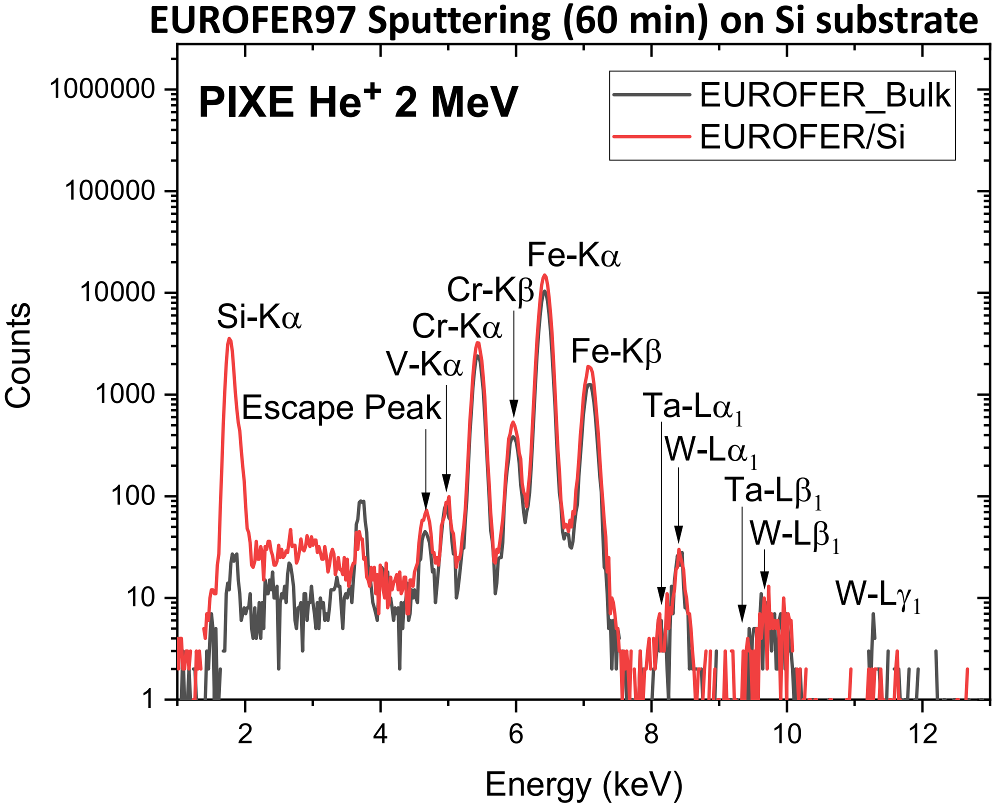


→ Homogeneous with low presence of contaminations.

[1] *KTH Royal Institute of Technology, Stockholm, Sweden*

W-P 5: Sputtering yields and BCA simulation

Formation and characterization of thin films from EUROFER97 target



PIXE analysis: identification of elements difficult to detect by RBS (V, Ta).

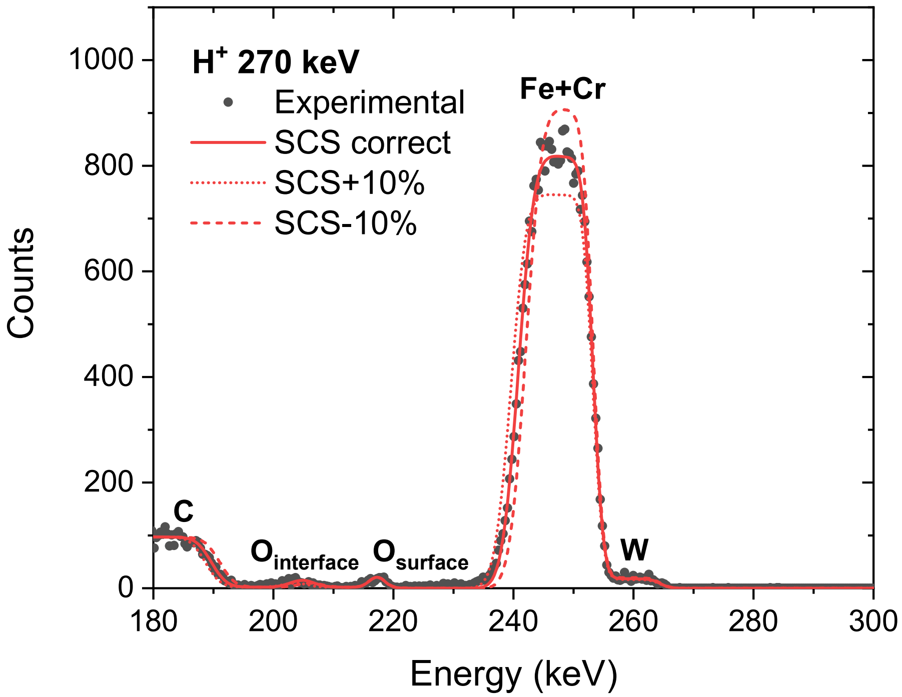
Investigation of material properties in comparison to bulk:
-Composition; Microstructure; mechanical properties; deuterium retention.

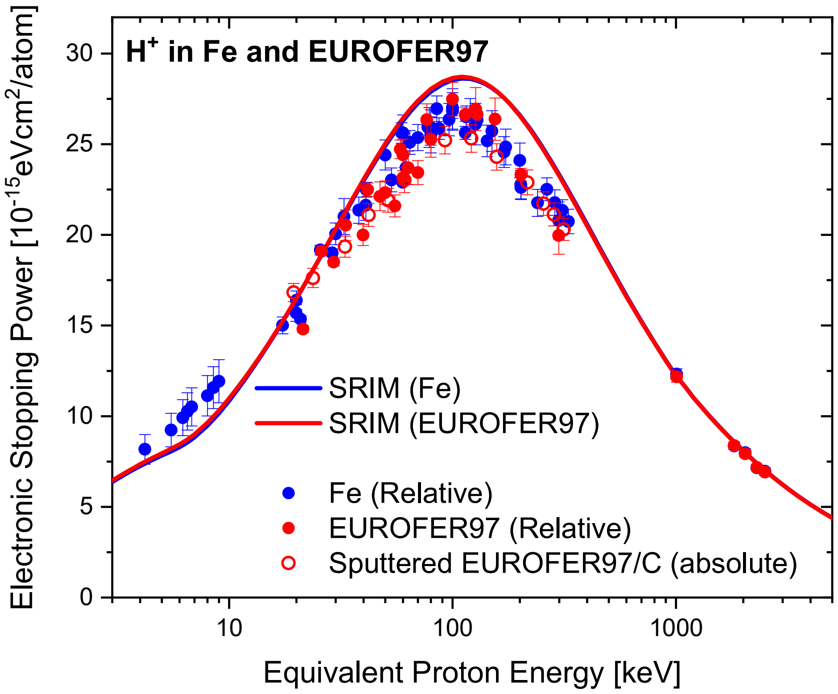
E. Pitthan, P. Petersson, T. T. Tran, et al.
Nuclear Materials and Energy **34** (2023) 101375.

W-P 3: Electronic energy loss measurements

Electronic SCS of thin films from EUROFER97 target

→ SCS based on the width of spectra in comparison to SIMNRA simulations (absolute approach).





→ Similar behaviour in comparison to relative approach (within 5%).

→ Similar SCS trend between bulk and redeposited EUROFER97.

W-P 5: Sputtering yields and BCA simulation

Sputtering yields and angular distributions of pristine W, Fe, and EUROFER97 samples.

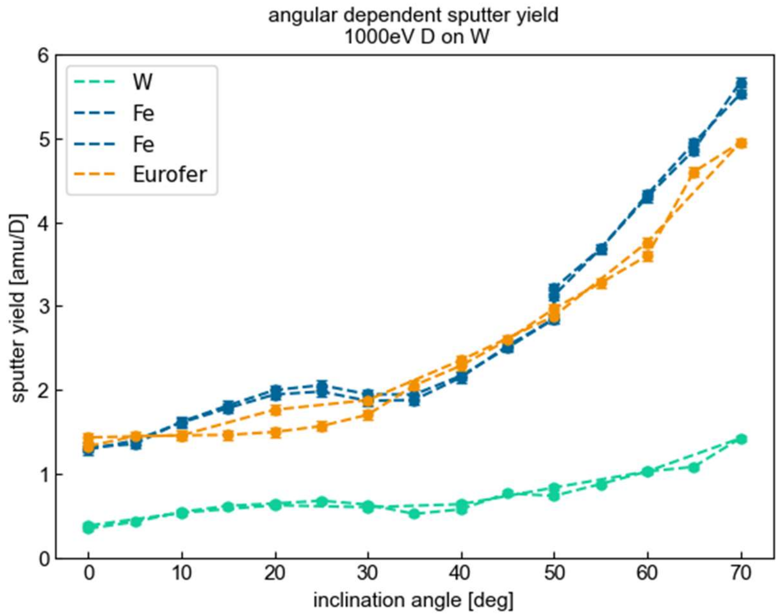
W and Fe films:

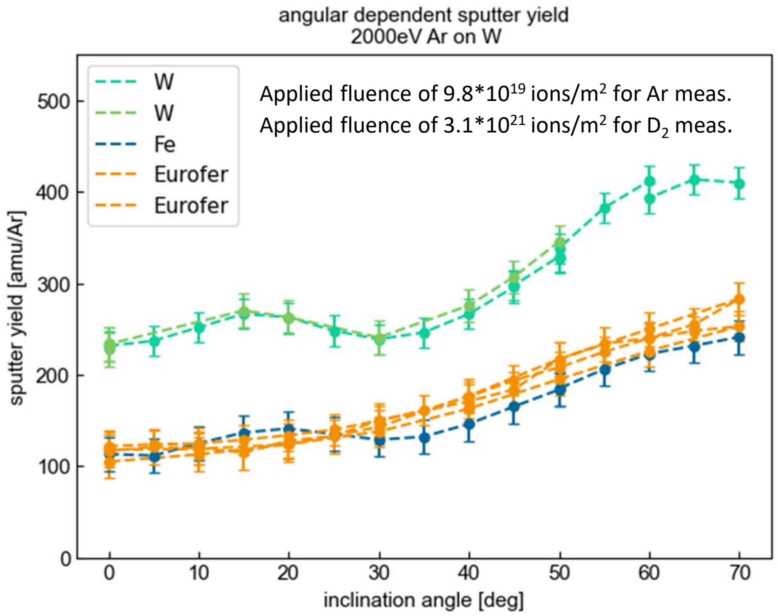
- local minimum at 30° - 35° found
- effect is pronounced for higher energies
- hypothesis: modified sputtering yield due **to crystallinity effects**

Films from EUROFER97:

- **no crystallinity effects** found
- similar sputtering yield values in comparison to Fe samples

→ **low fluences** do not suggest significant W surface enrichment during measurement.

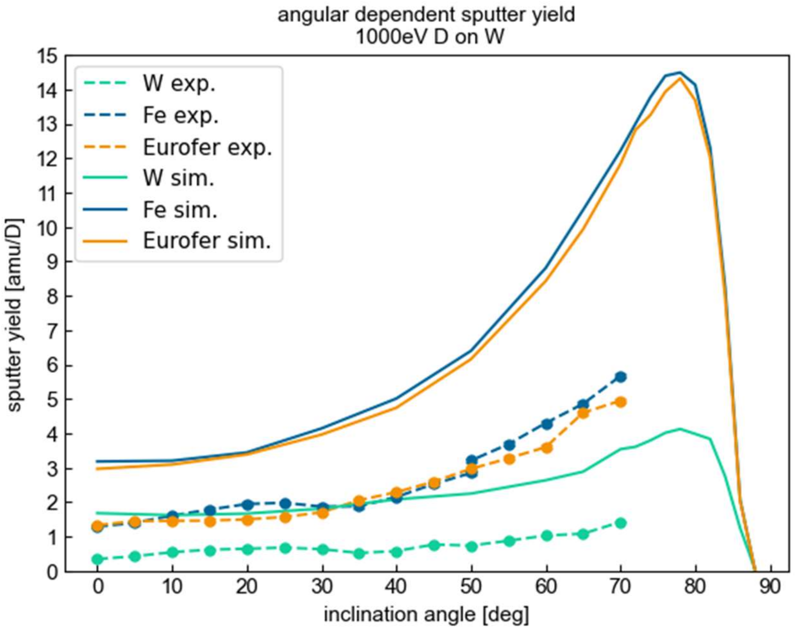

angular dependent sputter yield
1000eV D on W

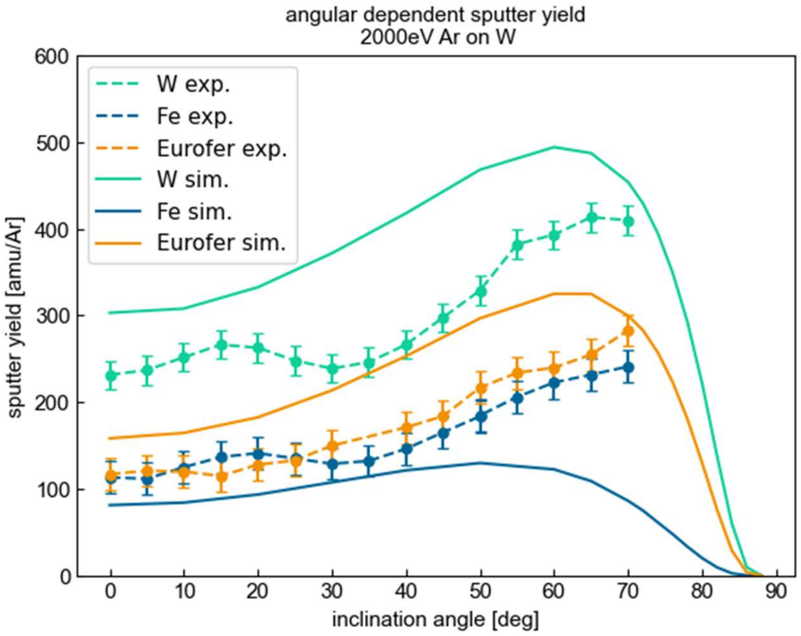

angular dependent sputter yield
2000eV Ar on W

Applied fluence of $9.8*10^{19}$ ions/m^2 for Ar meas.
Applied fluence of $3.1*10^{21}$ ions/m^2 for D_2 meas.

W-P 5: Sputtering yields and BCA simulation

BCA Simulations (SDTrimSP):

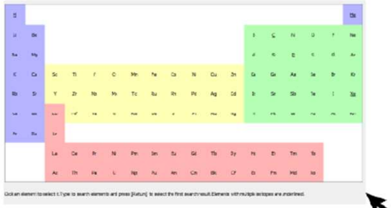




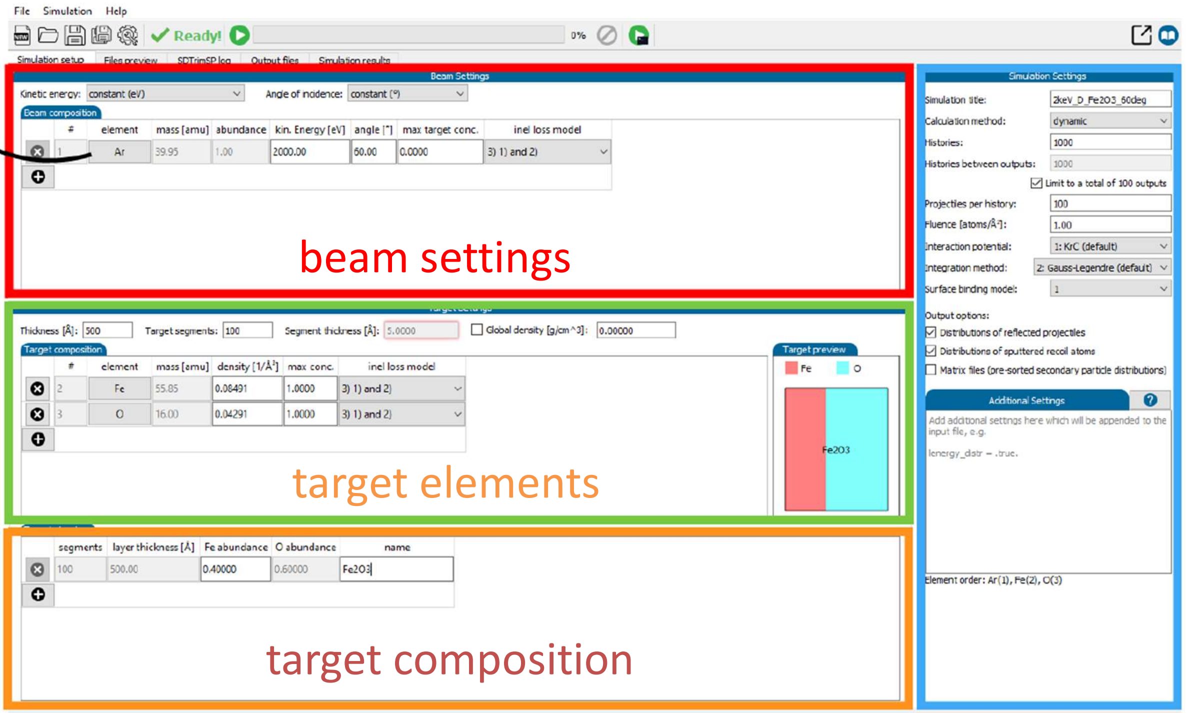
Simulations compared to experiments show general offset:

→ **Try different e-stopping models and interaction potentials used in the simulation.**

W-P 5: Sputtering yields and BCA simulation



Development of a SDTrimSP-GUI



Kinetic energy: constant (eV)
Angle of incidence: constant (°)

#	element	mass [amu]	abundance	kin. Energy [eV]	angle [°]	max target conc.	inel loss model
1	Ar	39.95	1.00	2000.00	60.00	0.0000	3) 1) and 2)

Thickness [Å]: 500
Target segments: 100
Segment thickness [Å]: 5.0000
☐ Global density [g/cm^3]: 0.00000

#	element	mass [amu]	density [1/Å³]	max conc.	inel loss model
2	Fe	55.85	0.08491	1.0000	3) 1) and 2)
3	O	16.00	0.04291	1.0000	3) 1) and 2)

segments	layer thickness [Å]	Fe abundance	O abundance	name
100	500.00	0.40000	0.60000	Fe2O3

Simulation title: 2keV_D_Fe2O3_60deg
Calculation method: dynamic
Histories: 1000
Histories between outputs: 1000
☑ Limit to a total of 100 outputs
Projectiles per history: 100
Fluence [atoms/Å²]: 1.00
Interaction potential: 1: KrC (default)
Integration method: 2: Gauss-Legendre (default)
Surface binding model: 1

Output options:
☑ Distributions of reflected projectiles
☑ Distributions of sputtered recoil atoms
☐ Matrix files (pre-sorted secondary particle distributions)

Add additional settings here which will be appended to the input file, e.g.

lenergy_distr = .true.

Element order: Ar(1), Fe(2), O(3)

special settings

(dynamics, statistics, potentials, extras)

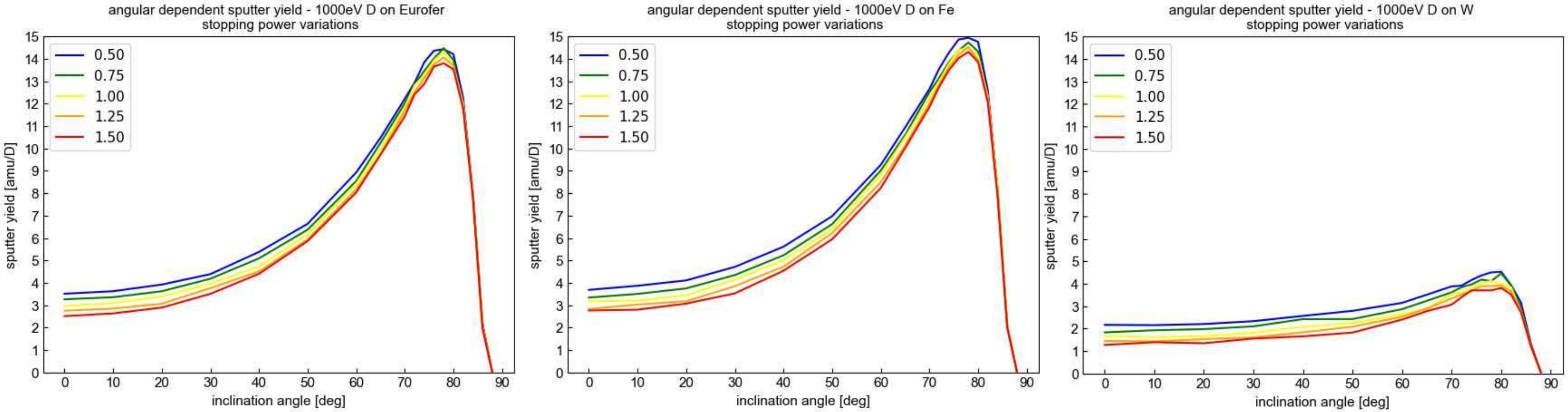
- easy-use (like SRIM)
- live visualisations
- Quick parameter sweep
- (e.g, potentials, binding energies,…)
- Other BCA codes soon integrated (TRIDYN, IMSIL)

https://doi.org/10.1016/j.nimb.2022.04.008

W-P 5: Sputtering yields and BCA simulation

Comparisons on the experimental sputtered yields between pristine Fe, W, EUROFER samples to Monte Carlo-based BCA simulations using input data from energy loss and interatomic potential.

Simulations using SDTrimSP-GUI (P. S. Szabo et al. Nucl. Instrum. Meth. B 2022).



In progress:

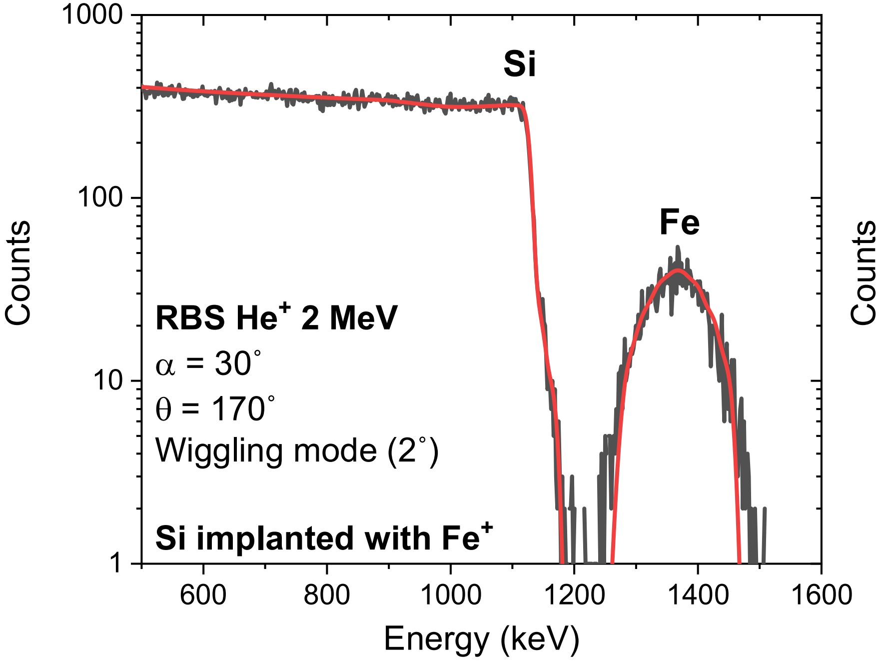
Improve output reading of SCS values.

Implement interatomic potential corrections for TFM.

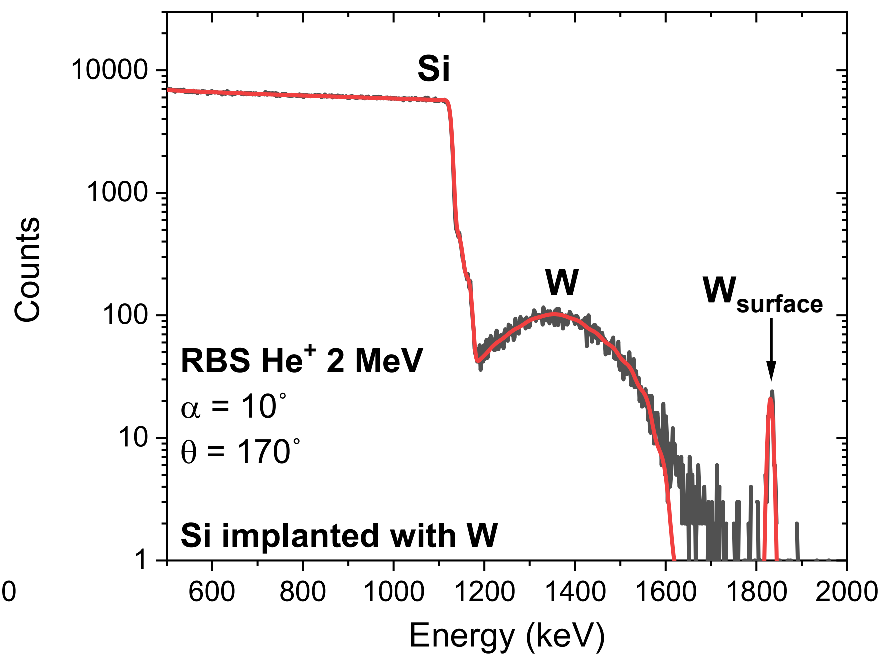
W-P 6: Ion-irradiation experiments

self-irradiation → *High levels of displacement damage without impurities.*
Ion Energy → *High levels of displacement damage within depth from SCS evaluation window.*

Fe^+ 300 keV → Fe, EUROFER, Si samples
Fe areal density: 42.25×10^{15} Fe/cm^2



W^{2+} 2.84 MeV → W and Si samples
W areal density: 1.57×10^{15} W/cm^2



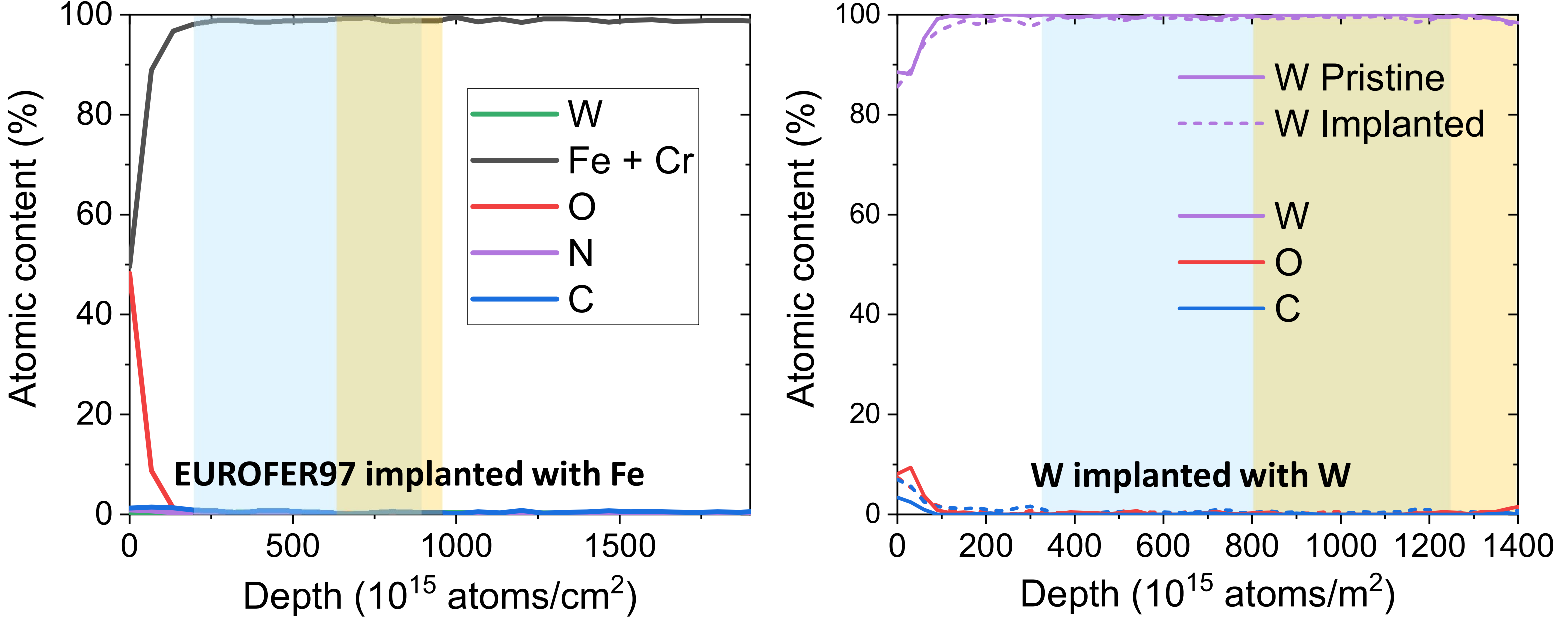
W-P 6: Ion-irradiation experiments

TRIM simulation:

→ Maximum damaged region

→ Ion range

ToF-ERDA (I^{8+} 36 MeV):



No significant modification on composition on self-irradiated samples.

Future steps: SCS and sputtering yield investigation using irradiated samples.

W-P 7: Computational modelling

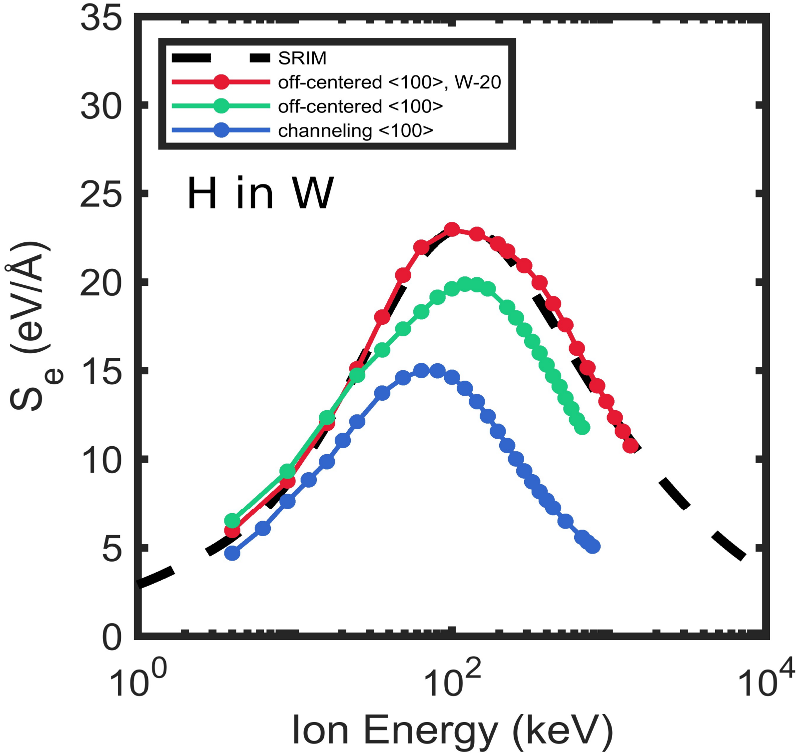
TDDFT electronic stopping power (D7.1)

- Electronic stopping power from rate of change of total energy of the system, with ion travelling at constant velocity
- Simulation cell: 3x3x4 supercell (72 W + 1 H atoms), lattice constant ***a***
- Qb@ll TDDFT code + LDA norm-conserving pseudopotentials.

- **Preliminary results (H on W):**
  Stopping power along <100> channel and off-center.

More simulations for this task are still being carried out (M7.1 target March 2023).

D7.2 effect of defects postponed to 2023.
D7.3 and D7.4 started earlier.



W-P 7: Computational modelling

Ion backscattering with MDRANGE (D7.3)

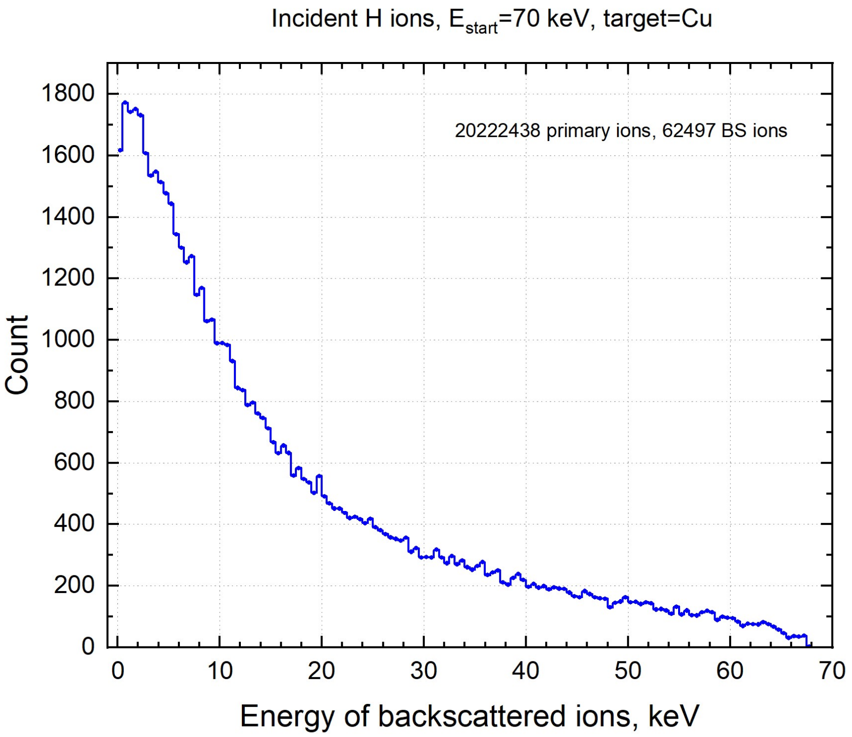
Original task description:
D7.3: MDRANGE simulations of ion implantation ranges and sputtering yields from surfaces with evolving composition.
Started earlier than originally planned.

Task goal modified to compute instead backscattered ion energy spectrum to better correspond to Uppsala experiments,
aim: detailing electronic and nuclear energy deposition.

Status:
Modification of MDRANGE for backscattering calculations completed
Data for energy spectrum of backscattered ions obtained for 70 keV H on Cu.
To do: filter for scattering angle.

Future work:

- Include TDDFT-calculated electronic stopping (obtained in D7.1 and D7.2)
- Experimental ion energies and targets
- Statistics: >1M ions for each case



W-P 7: Computational modelling

Sputtering with MD simulations (D7.4)

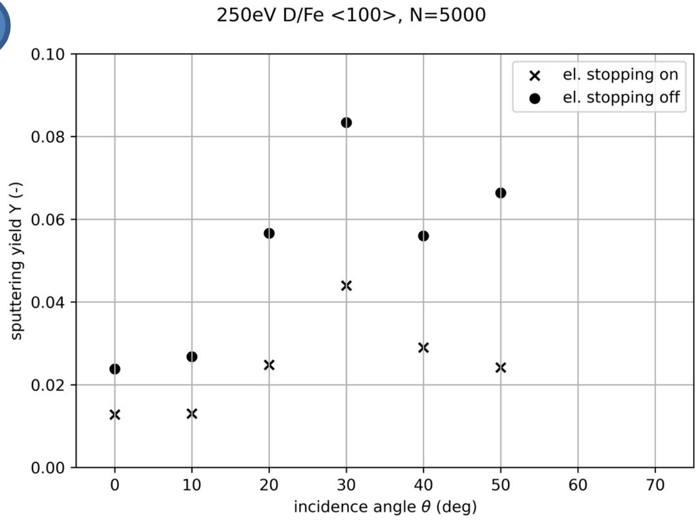
- Classic MD with el. stop. as friction force from SRIM 2013, shown to affect sputtering (**plot A**)
- Simulation cell: ~16k Fe + 1 D atoms, equilibrated at 300K
- LAMMPS MD code + EAM interatomic potential
- N=5000 Monte Carlo-style simulations per angular step, varying ion azimuthal angle and impact point

Preliminary results (250eV D on Fe):

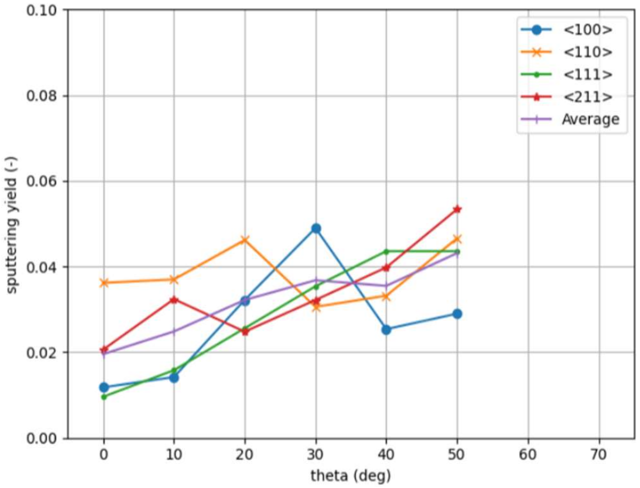
Plot B: dependence of sputtering yield on ion incidence angle, for 3 low-index orientations (<100>, <110>, <111>, (<211>)) in pristine crystal.

- Ongoing work:
  - Uneven surface (step defect, irradiated).

A



B



Summary

Deliverables for 2022:

W-P 1: General management

D1.2 → Two annual meetings.

W-P 2: Sample preparation

D2.6 → Routinely performed quality control cross-checks.

W-P 3: Electronic energy loss measurements

D3.2 → Stopping power of pristine PFCs samples for light ions (sub-keV regime).

W-P 4: Interatomic potential measurements

D4.1 → In-situ preparation of the pristine crystalline Fe and W samples for ToF-LEIS/MEIS.
D4.2 → ToF-LEIS/MEIS angular scans measurements (pristine samples).

W-P 5: Sputtering yield & QCM

D5.2 → Sputtering yields and angular distributions of pristine W and EUROFER97 samples.

W-P 6: Ion-irradiation experiments

D6.1 → Define irradiation conditions: ions, energy, fluencies within different (ex-situ and in-situ) set-ups.
D6.2 → Ex-situ ion irradiation experiments on the PFCs samples at UU.

W-P 7: Computational modelling

D7.2 → Theoretical calculation of electronic stopping power for random trajectories of light ions in W, Fe and Fe-alloys containing defects, using TDDFT. (postponed: D7.3 and D7.4 started earlier).

Summary

Milestones achieved:

2021:

M2.1 → Sample characterization: pristine samples.
M3.1 → Experimental energy loss results from pristine PFCs samples (keV regime).
M5.1 → QCM set-up assembling at UU.

2022:

M2.2 → Sample characterization: damaged samples.
M2.3 → Periodically quality control cross-checks.
M3.2 → Experimental energy loss results from pristine PFCs samples (sub-keV regime).

Partially:
M5.1 → Comparisons on the experimental sputtered yields between pristine Fe, W, EUROFER samples to Monte Carlo-based BCA simulations using input data from energy loss and interatomic potential.
M7.1 → Ab-initio calculations for energy loss and comparison to results: pristine samples.
M7.3 → Implementation of geometrically dependent electronic energy losses in the MDRANGE ion range code.

Summary

Future milestones (2023 & 2024):

M3.3 → Experimental data from damaged W & Fe.
M3.4 → Experimental data from damaged EUROFER.

M4.1 → Determination of short-range interactions from experimental spectra and BCA calculations (in progress).
M4.2 → Comparison of the short range interactions for pristine vs. damaged samples: defects influence.

M5.2 → Benchmarking sputtering yield codes with input data from WP3 and WP4 (in progress).
M5.3 → Evaluate how sensitive the sputtering yield is in terms of energy loss and interatomic potential when local defects and impurities (i.e. damaged samples) are present.

M6.2 → Depth profile of the irradiated Fe, W and EUROFER samples suitable for WP4.

M7.2→ Ab-initio calculations of damaged samples and comparison to experiments: local effects on the fundamental quantities.
M7.4 → Comparisons of predicted and experimentally measured electron energy losses for input into MD based simulations (in progress).
M7.5 → Experimentally deduced short range interactions as inputs for MD based simulations of sputtering yield (in progress).

Extras