ANNUAL ION BEAM FACILITY REPORT 2021

DIFFER

Annual
Ion Beam Facility
Report

2021

Annual Ion Beam Facility Report 2021

This report summarizes the activities at the DIFFER Ion Beam Facility (IBF) in 2021. The accelerator was used for 936 hours within 119 days to conduct research (100% ion beam analysis) and 115 days were spent on mainly repairs but also maintenance and implementation of lab improvements such as the magnetic compensation system or CODAC. The report gives a short summary of the academic research and performed repairs/maintenance.

Facility Manager: Dr. Hans van Eck
Project Leader: Dr. Beata Tyburska-Pueschel
Accelerator operator: Wim Melissen
Radiation safety: Santi Alonso van den Westen

https://www.differ.nl/research/fusion-facilities-and-instrumentation/ion-beam-facility
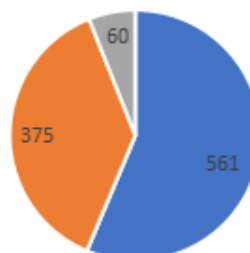
De Zaale 20
5612 AJ Eindhoven, The Netherlands

IBF in numbers

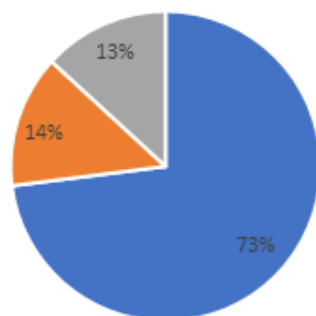### IBF usage days in 2021
(during 251 working days)

78
41
115
17
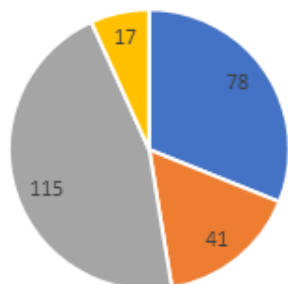
■ DIFFER ■ Industry ■ Repairs ■ Un-used

Accelerator usage hours in 2021

60
375
561

■ DIFFER ■ Industry ■ Other

Ion beam usage in 2021

13%
14%
73%

■ 4He ■ 3He ■ 1H

Accelerator users in 2021

1
2
1
4
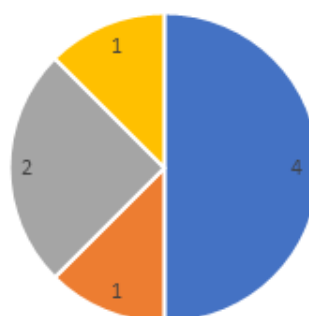
■ Fusion ■ Industry ■ Solar Fuels ■ TU/e

IBA chamber usage in 2021

0%
14%
86%
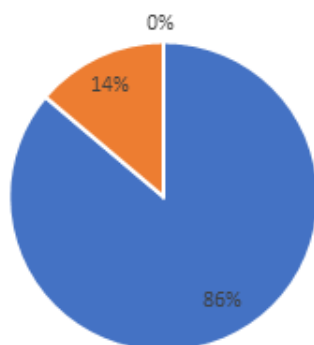
■ IBAS ■ TEAC ■ UPP

Facility description

The IBF housed by DIFFER is currently the only such facility available in the Netherlands. A 3.5 MV Singletron from HVEE is the pillar of the IBF and it is capable of producing any gas ions, with present use focusing on H, 3He, and 4He. The advantage of this type of accelerator is high beam stability with low ripple and high beam current. The accelerator has 7 beamlines, of which 3 are in use: one is linked to Magnum-PSI allowing for the world's highest plasma-flux exposure and for in-situ IBA (TEAC chamber), the second leads to UPP allowing for in-operando IBA and mid-flux plasma exposure (under construction), and the third goes to an ex-situ IBA chamber called IBAS.
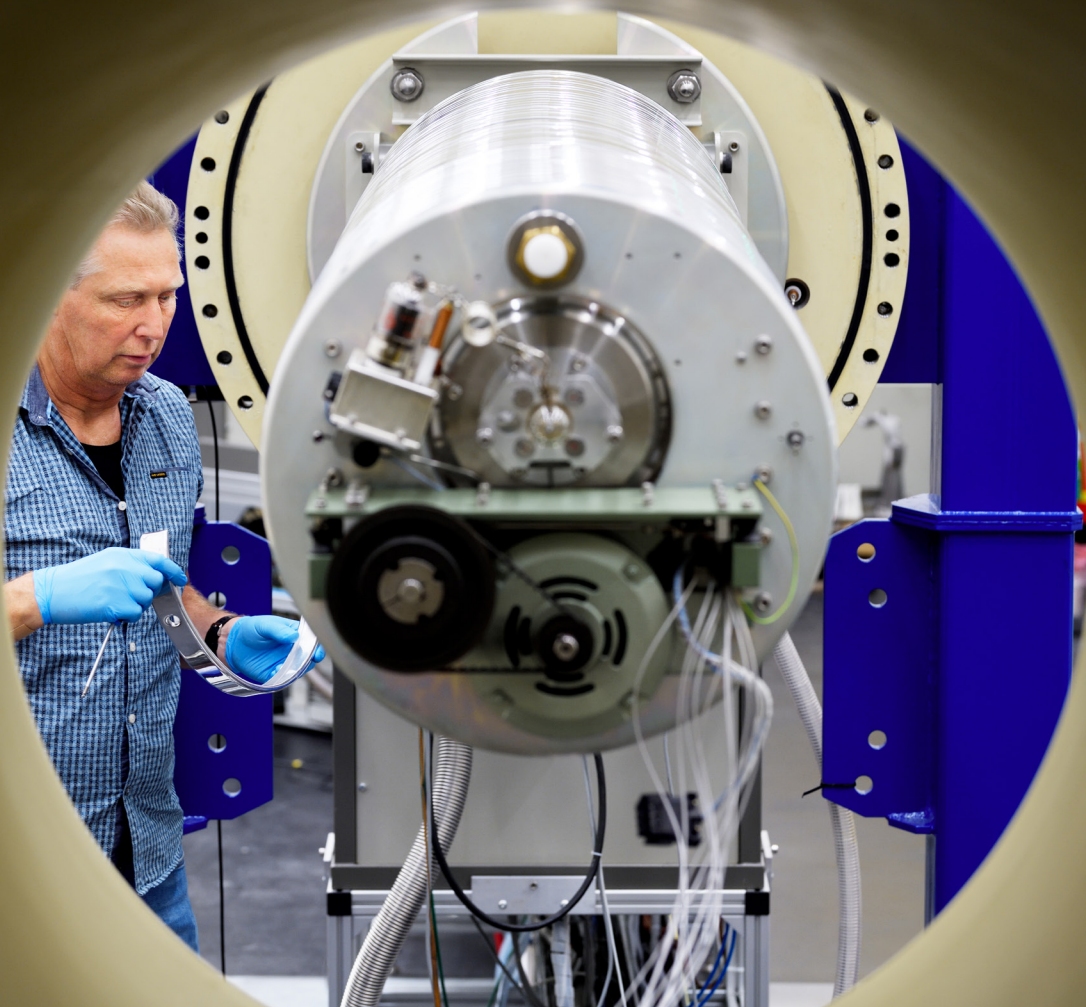
Plasma-induced deuterium uptake of EUV mirrors

S. Wang and T.W. Morgan (DIFFER FE)

Blister formation in thin films have recently been an emerging research subject in relation to extreme ultraviolet (EUV) mirrors in lithography tools. This device contains hydrogen plasmas and Sn droplets. The Ru-capped EUV mirrors are exposed to high-flux ions and radicals coming from the plasmas. Blister formation results from hydrogen atoms retained in the materials and could lead to delamination of thin films. We experimentally studied the effect of Sn on Ru-capped targets exposed to deuterium plasmas.

We found a strong correlation between the deuterium uptake and the amount of Sn on the Ru surface. As shown below, the deuterium content in the target dramatically increased with the amount of Sn on the target surface. This showed that Sn on the surface accelerates deuterium uptake of the target. The result is qualitatively consistent with recent work by Onwudinanti et al., who showed the presence of Sn on Ru lowers the energy barrier for hydrogen to move from the surface to the sub-surface

Reference: C. Onwudinanti, I. Tranca, T.W Morgan, S. Tao, Tin, the enabler - Hydrogen diffusion into ruthenium, Nanomaterials 9 (2019) 129
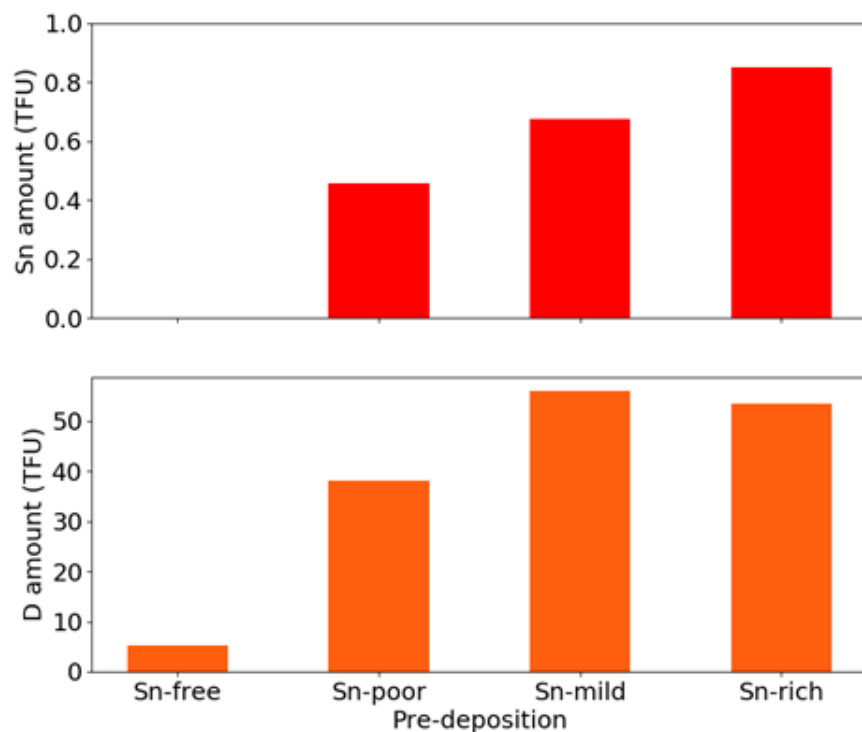

Figure: Sn and D contents of Ru-capped targets exposed to deuterium plasmas. The measurements were performed using Rutherford backscattering spectrometry (RBS) and elastic recoil detection (ERD) at DIFFER ion beam facility

Deuterium retention in liquid metal

W. Ou, T.W. Morgan, W. Arnold Bik, P. Rindt, R.S. Al, J.W.M. Vernimmen, S. Brons (DIFFER FE)

Liquid metals (LMs) such as liquid tin (Sn) and lithium (Li) have been proposed as plasma-facing materials (PFMs) in future fusion reactors to solve those issues that can be deadly to solid materials, such as heat exhaust and neutron radiation damage. Compared with solid materials, LMs are capable of withstanding high heat fluxes by evaporation and vapor shielding and they can also refresh and self-heal through flow, protecting underlying substrate materials. Due to their liquid nature, LMs could be immune to most of negative effects of neutrons. While, tritium (T) inventory in LMs under fusion-relevant plasma conditions is still unclear and remains largely unexplored. High T inventory in PFMs could pose safety concerns as T is radioactive. Additionally, T is scarce and a precious fusion fuel. A high fuel efficiency is required for a commercial nuclear fusion station.

In this work, we used deuterium (D) as a proxy of T to explore D retention in Sn and Li, which are two of most promising candidates for liquid metal PFMs, under fusion-relevant flux plasmas that can be simulated on Magnum-PSI.

Liquid Sn and Li were reserved in capillary porous system (CPS) samples either consisted of molybdenum meshes with 104 μm pores or made of 3D-printed tungsten with porous textures and exposed to D plasma on Magnum-PSI at high flux 1023 ~ 24 m-2s-1. Nuclear reaction analysis (NRA) was applied to characterize D retention in top surface. It's a non-destructive technique to quantity the concentration of those selected light elements that can have nuclear reaction with the incident high energy ion beam. NRA was performed on IBF set-up at DIFFER, which is directly connected to Magnum-PSI, making it possible to immediately measure D depth profiles in the samples after plasma exposure without breaking vacuum. This is extremely significant for characterizing D retention in Li as it is highly reactive in the air. In this work, 2.5 MeV 3He+ beam was used following the reaction 2D(3He, p)4He and has a probing depth around 6.7 μm for Sn and 32 μm for Li . An example of typical D depth profiles in Li after D plasma exposure can be found in Fig. 1 (a). Thermal desorption spectroscopy (TDS) was also used to collect total D retained in the whole samples.
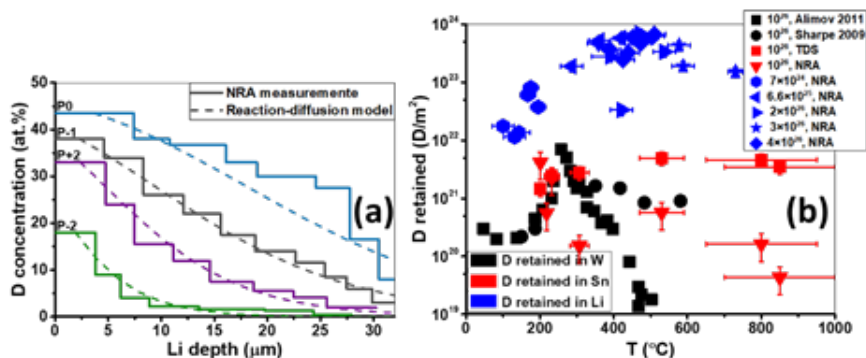

Figure 1 (a) D concentration depth distribution at different position on a Li sample. Solid lines are NRA measurement results; dash lines are the modelling from reaction-diffusion model. (b) A comparison of D retention among tungsten, tin and lithium as a function of temperature at different D+ ion fluence

A general conclusion of this work is that D retention in Li is extremely high which can be 2 ~ 4 orders of magnitude larger than that in W due to LiD formation while D retention in Sn is comparable to that in W at low temperature but 1 ~ 2 orders of magnitude higher when temperature is high, which is most likely due to D_2 gas bubble formation. A comparison of D retention in W, Sn and Li is presented in Fig. 1 (b).

In-situ LIBS D concentration measurements and comparison with NRA in W, W-Ta(5%) and W-O coatings exposed to linear plasma device Magnum-PSI plasma

I. Jõgi, P. Paris, K. Piip, H. van der Meiden, J. Vernimmen, B. Tyburska-Puschel (DIFFER FE)

We have investigated the applicability of Laser Induced Breakdown Spectroscopy (LIBS) for analyzing the changes in the composition and fuel retention of W coatings on Mo substrate (5 μm W+5%Ta, 5 μm W, ~1 μm porous W, ~1 μm columnar W) following exposure to D_2 or mixed D_2-N_2 plasma beams in the linear plasma device Magnum PSI. The LIBS data was compared with D retention and depth profiled measured by Nuclear Reaction Analysis (NRA).

Porous W on Mo is fully oxidized with a thick WO/WO_2 formation - porous coatings show uniform D concentration and oxide formation throughout the entire sample (Fig. 2). Other coatings oxidize mainly in the ring center (plasma exposure ring) and the oxide thickness is much thinner –

none-porous coatings show the highest oxidation and the lowest D retention in the sample center (Fig. 1). N-seeding has no effect on D retention but it might promote oxidation as oxide thickness is on average bit higher after D+N exposure.

The measured coating thickness after plasma exposure is comparable to manufactured. Generally, the substrate roughness is below 10˚ but it increases locally above 10˚, which suggests that the coating partially flaked off and plasma exposure caused substrate sputtering. The flaking occurs in- and outside the ring.

The LIBS retention and D depth profiles show good qualitative agreement with NRA. Quantitative agreement varies strongly between measurement points. W-Ta(5%) coating seemed to have 50% higher D/W ratios (not seen by NRA).
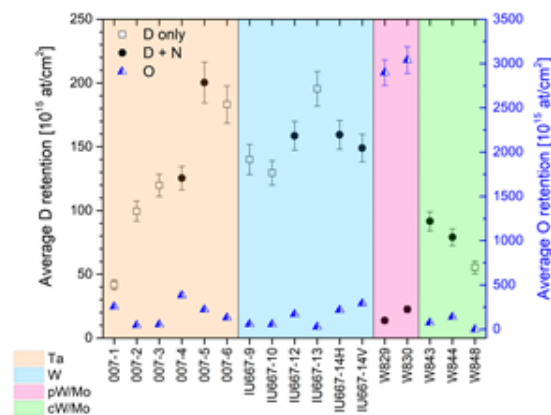

Fig 1: D retention in coatings: 5 μm W+5%Ta (orange), 5 μm W (blue), ~1 μm porous W (pink), and ~1 μm columnar W (green)
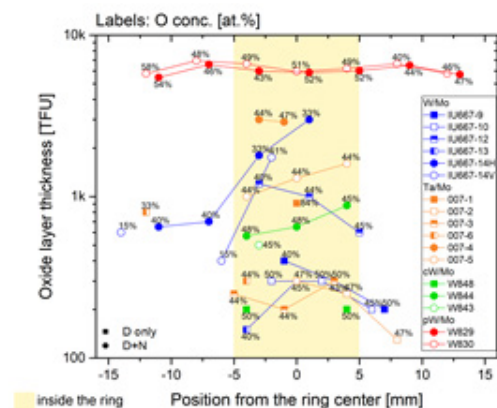

Fig.2: Oxide formation inside and outside the plasma exposure ring in various coatings (fig.1 colors apply). Labels show amount of oxygen.

Suppression of hydrogen blistering in Mo/Si layered structures

V. Vollema and J.W.M. Frenken, Advanced Research Center for Nanolithography (TUe)

Hydrogen blistering is receiving renewed attention in the scientific community due to its appearance in technologically advanced areas such as nuclear fusion and extreme ultraviolet lithography (EUVL). In the latter case, EUV mirrors consisting of stacks of alternating Mo and Si layers are continuously subjected to hydrogen radicals and low energy hydrogen ions, which can result in the accumulation of hydrogen atoms within the structure. Such accumulation can potentially lead to recombination of hydrogen into (gas) molecules, clustered in the form of gas pockets: blisters. These blisters can be expected to reduce the quality of the refined mirror structure. Interestingly, the precise mechanisms by which this process starts are yet to be revealed.

In this context, we investigated the effect of the deposition parameters during manufacturing of an Mo/Si layered structure on the proneness to hydrogen blistering of such a structure. We found that modest variations in one of them could make the difference between severe hydrogen blistering or none at all (see fig. 1). Using ERD, we verified that this was correlated with a significant difference in hydrogen retention in the two cases. The ERD spectra show that hydrogen uptake resulting from deliberate hydrogen ion exposure is almost one order of magnitude higher for the structure that blisters than for the blister-resilient structure. This increase in hydrogen signal in the blistering case can be explained either by the accumulation of hydrogen gas in the blisters, or by a lowered out-diffusion of implanted hydrogen. Additional experiments point towards the former and further experiments are in progress to verify whether in-plane mechanical stress in these layers and their change due to incorporation of hydrogen play a key role in blister nucleation.
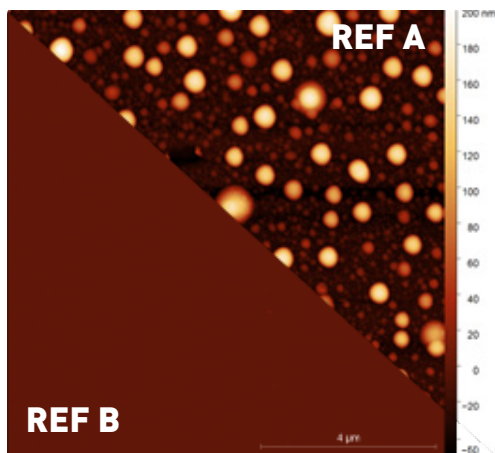

Figure 1: AFM images of two differently deposited Mo/Si layer structures after fully equivalent hydrogen ion exposures, displayed on the same color-height scale. One structure severely blisters (top right), while the other does not degrade at all (bottom left).
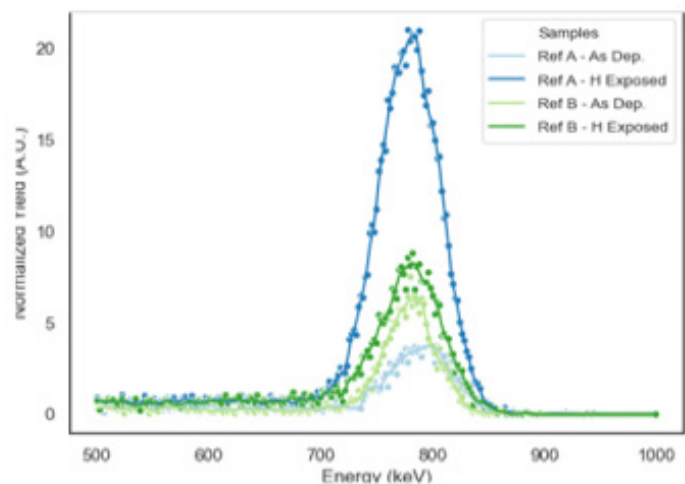

Figure 2: The hydrogen peak of ERD spectra for the two differently manufactured sample types, both before and after exposure. The increase in hydrogen content due to exposure is very modest for the blister-resilient structure (green) compared to the blister-prone structure (blue).

BiFeO$_3$ thin films for photoelectrochemical water splitting

Nitin P Prasad (DIFFER SE)

BiFeO$_3$ has been long identified as a potential photo(elec-tro)catalyst due the presence of an inbuilt ferroelectric field that can aid charge separation and prevent recombination.[1] Introducing excess Bi in the synthesis of BFO often leads to favorable improvements like lower leakage current, better ferroelectricity and reduced Bi vacancies[2]. Such changes can have varying effect on the photoelectrochemical performance of BFO. This study focusses on the effect of Bi-Fe stoichiometry on BiFeO$_3$ thin films photocathodes for hydrogen evolution reaction.

BFO thin films were synthesized by sol gel technique. The precursor solution with two different Bi-Fe stoichiometry (stochiometric, 10% excess Bi) was spin coated on fluo-rine doped tin oxide. The Ion Beam Facility (IBF) was used primarily for confirming the composition of the thin films. Reports from literature suggest that a thin Bi rich layer is formed on annealing BiFeO$_3$ thin films. Thus, by annealing the thin films after each cycle, there is a possibility of the formation of a stacked structure. The main goal of using the IBF was to see if we could identify this layered structure by looking at depth profile of Bi-Fe composition. For this purpose, Rutherford Backscattering Spectroscopy (RBS) was used. Results suggested that a Bi rich layer could be seen for films with two spin coat cycles. However, for films with 4 layers, this was not observed. Nevertheless, the compositions of the thin films were confirmed with the Bi-Fe ratio for stoichiometric thin films found to be 0.97 while that for the 10% excess Bi thin films to be 1.07. Photoelectrochemical studies showed that excess Bi thin films have a higher activity for HER as compared to the stoichiometric thin films. This is likely due to the reduction in Bi vacancies on adding excess Bi.

References

[1] Cao, D.; Wang, Z.; Nasori; Wen, L.; Mi, Y.; Lei, Y. Switchable Charge-Transfer in the Photoelectrochemical Energy-Conversion Process of Ferroelectric BiFeO3 Photoelectrodes. Angewandte Chemie - International Edition 2014, 53 (41), 11027–11031. https://doi.org/10.1002/anie.201406044.
[2] Lahmar, A.; Zhao, K.; Habouti, S.; Dietze, M.; Solterbeck, C. H.; Es-Souni, M. Off-Stoichiometry Effects on BiFeO3 Thin Films. Solid State Ionics 2011, 202 (1), 1–5. https://doi.org/10.1016/J.SSI.2011.03.017.
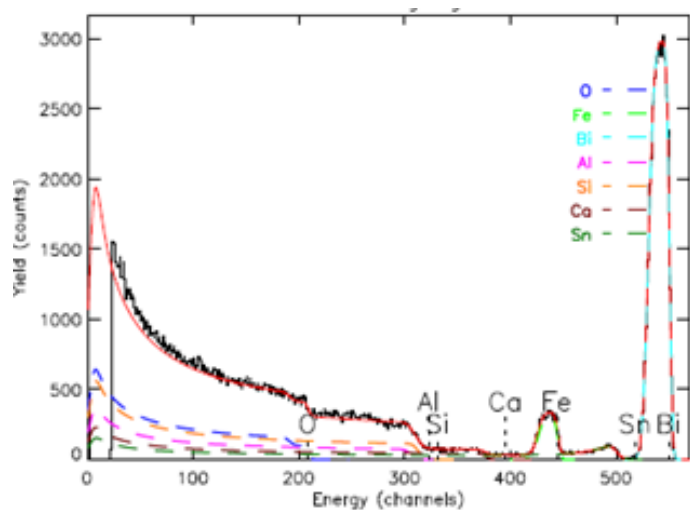
Figure 1 : Sample RBS spectra recorded for a stoichiometric thin film of BiFeO$_3$ on glass substrate

Determination of retention levels during lithium and deuterium co-deposition in Magnum-PSI

M. Morbey, W. Arnold Bik , B. Tyburska-Pueschel , E. Zoethout, S. Wang, and T. W. Morgan (DIFFER FE)

We investigated the deuterium retention in lithium and deuterium co-deposition. A CPS (capillary porous structure) made of W was filled with lithium and exposed to deuterium plasma B=0.8T, I=100A and 4.5 slm D for 100s. Six 1x1cm SS plates were placed close to the CPS in order to collect the lithium and deuterium coming from the target and the plasma, respectively. These SS plates were placed on a heater and the temperature varied between room temperature and 400°C. NRA and EBS were performed one after the other. NRA was the first choice as it has been used in similar experiments before. However, the compromised Li cross sections made the data hard to interpret. Therefore, EBS was performed as well. The lithium content can easily be deduced from the EBS spectrum but the D content due to its light mass and the heavy substrate (Fe) is harder to extract. For NRA 2.3MeV 3He+ was used, while for EBS 2.1 MeV protons beam was chosen. The probing depth for the NRA is around 15.6 um for LiD using a density of 0.8 g/cm3 and the EBS is 104.25 um. From FIB and SEM it was observed that the thickest layer was around 3.4um. The analysis of these spectra is not finished yet but from area content of the Li peak in EBS and NRA D peak we see no influence of the temperature in the Li/D ration neither of the distance to the CPS target. Three different experiments were done with the heater at 400°C and in one of them (the first one) almost no Li or D was detected. A QMC (quartz microbalance crystal) was used to monitor the lithium evaporation rate but we only have reliable data for the first and last 400°C experiment. We see double the evaporation (on average) for the last 400°C experiment. This does not explain the orders of magnitude difference in the lithium content of the WPs. Heater tests were performed and it was seen a 50°C different between the hottest and coldest witness plate. However outside the test we can only read one temperature. This might lead to temperature differences in the 400°C cases. It is possible that around 400°C we are in a transitional regime.
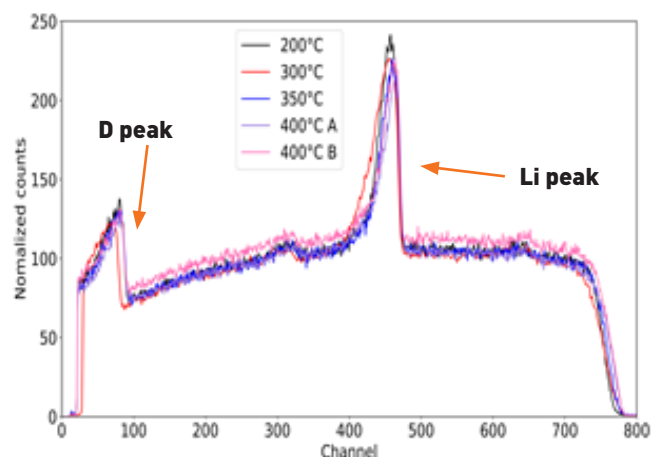

Figure 1. Typical EBS spectrum of a Fe substrate with a Li and D peak for WP1, the closest to CPS with different T. For the first 400°C experiment there is no EBS spectrum. No clear dependence on the temperature is seen. The 300°C case is rescaled for easier comparison since the amplification of the detector was changed between 300°C and all the other experiments
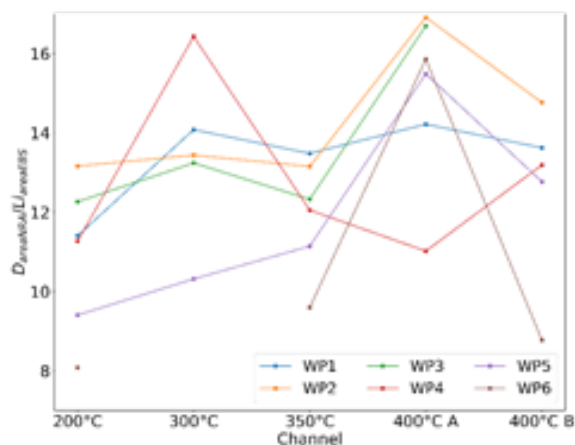

Figure 2. Ratio of the area of the deuterium peak from the NRA spectrum and the area of the Li peak from EBS. We do not see a clear dependence on temperature or distance to the plasma.

Determination of the iridium loading of catalyst coated polymeric membranes

M. Tsampas and B. Tyburska-Puschel (DIFFER SF)

Polymer electrolyte membrane (PEM) water electrolysis is a key technology for a sustainable hydrogen economy, but costs still have to be reduced to be competitive with hydrogen production from fossil resources. Currently, bipolar plates and porous transport layers make up for more than half of the PEM water electrolyzer stack costs. However, with decreasing costs for these components the noble metal based catalysts necessary for high-conversion rates and efficiencies become a major cost driver The platinum required for the hydrogen evolution reaction can generally be reduced to values below 0.1 mg/cm2 without significantly losing performance. In contrast, reducing the amount of iridium for the oxygen evolution reaction remains a key challenge for PEM water electrolysis.

The Ir loading is conventionally determined by (i) weighing a reference substrate in a high-precision scale and correcting to the pure iridium content, and (ii) ICP-MS (inductively coupled plasma-mass-spectrometry) were the samples are dissolved in acid. The latter is not very efficient method for Ir catalyst-coated membrane because the substrates are very stable in acidic conditions. Thus IBA analysis was employed as an alternative technique, which apart from the Ir loading you can also determine the amount of oxygen in the IrOx films.

Several IrOx coated membranes were examined by RBS (2.4 MeV 4He) to determining the Ir content and the Ir to O ratio. The Ir concentration varies between 20 and 46 at.% and in some membranes, it decreases with the depth.

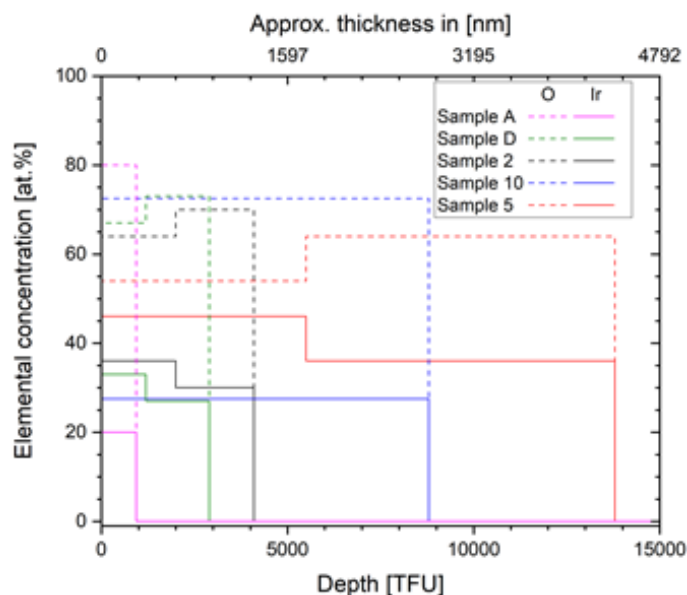| Sample name	Ir content [mg/cm^2]
Sample A	0.06 ± 7%
Sample D	0.274 ± 6%
Sample 2	0.43 ± 6%
Sample 10	0.77 ± 3.5%
Sample 5	1.76 ± 8%

Tab.1. Ir content in various samples.

Fig.1. Ir and O depth profiles in five different samples.

List of repairs in 2021

2.	Source replace, source defect	January
3.	Start-up source problem, bad contact	February
4.	Pump LEYBOLD D16B defect, low oil	February
5.	Start-up source problem, SF6 in source	February
6.	SF6 leak in vacuum system, various leaks	February/ March
7.	TML leaking	March
8.	RF BOX replace, RF box defect	March
9.	Vacuum leak gas system, various leaks	March
10.	VS1 TURBO pump has failed	May
11.	Still very small SF6 leak in the vacuum	May
12.	Water leak near 90° magnet	April
13.	PFEIFFER MAXIGAUGE TPG256A is defective	June
14.	Source replace, source defect	July
15.	RF tube in RF box defect, 2x tube replaced	August
16.	PLC defect, new PLC install	September

Publications

1. W. Ou et al., "Deuterium retention in Sn-filled samples exposed to fusion-relevant flux plasmas", Nucl. Fusion 60 (2020) 026008
2. W. Ou et al., "Deuterium retention in liquid lithium determined by in-situ NRA in Magnum-PSI", to be submitted to Nucl. Fusion (2021)
3. W. Ou, "Liquid metal as the wall material of fusion reactors: deuterium retention and surface stability", Ph.D. thesis 2021
4. I. Jõgi et al., "LIBS applicability for investigation of re-deposition and fuel retention in tungsten coatings exposed to pure and nitrogen-mixed deuterium plasmas of Magnum-PSI", Phys. Scr. 96 (2021) 114010