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Retention and permeation of hydrogen isotopes through RAFMs

Outline

- Institutions involved in our research, their abilities
- Motivation for this research in Russia
- What is already done by our group
- Our plans in frames of CRP

Institutions involved in our research

NRC 'Kurchatov Institute', Moscow, Russia
- coordination
- exposure in gas and plasma
- permeation experiments

TRINITI, Troitsk, Russia
- exposure to high heat fluxes
- exposure to high density plasma

A.A. Bochvar Institute of Inorganic Materials, Moscow, Russia
- producing RUSFER
- testing physical & mechanical properties
- exposure to T

National Research Nuclear University "MEPhI", Moscow, Russia
- retention analysis

Max-Planck-Institut fuer Plasmaphysik, Garching, Germany
- retention analysis

Persons involved in our research

N.P. Bobyr, D.I. Cherkez, A.V. Golubeva, B.I. Khripunov,
A.A. Mednikov, A.V. Spitsyn

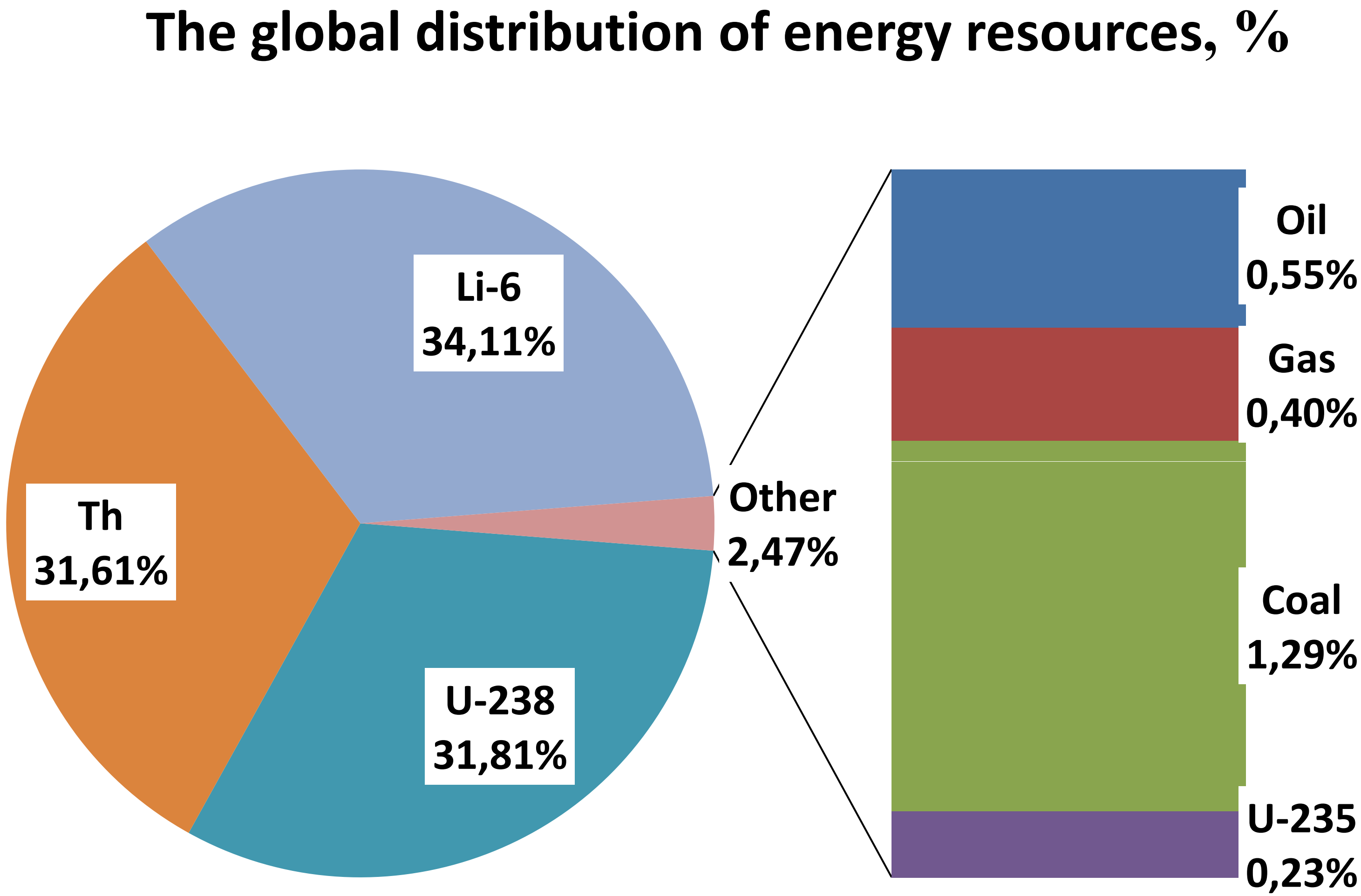
N.S. Klimov, A. Putrik

V.M. Chernov, I.G. Lesina, B. Ivanov

Yu. M. Gasparyan, V.S. Efimov

M. Mayer, M. Zibrov

Motivation: Energy resources



- Fission has highest potential and can cover all needs of humanity
- It is very difficult to exclude use of Fission energy in future

Motivation: Hybrid reactor project in Russia

Idea to use Fusion neutrons for fission was published by Igor Kurchatov in 1951
Since 1970s hybrid systems for nuclear fuel breeding and incineration of long life isotopes are discussed in Russia and USA

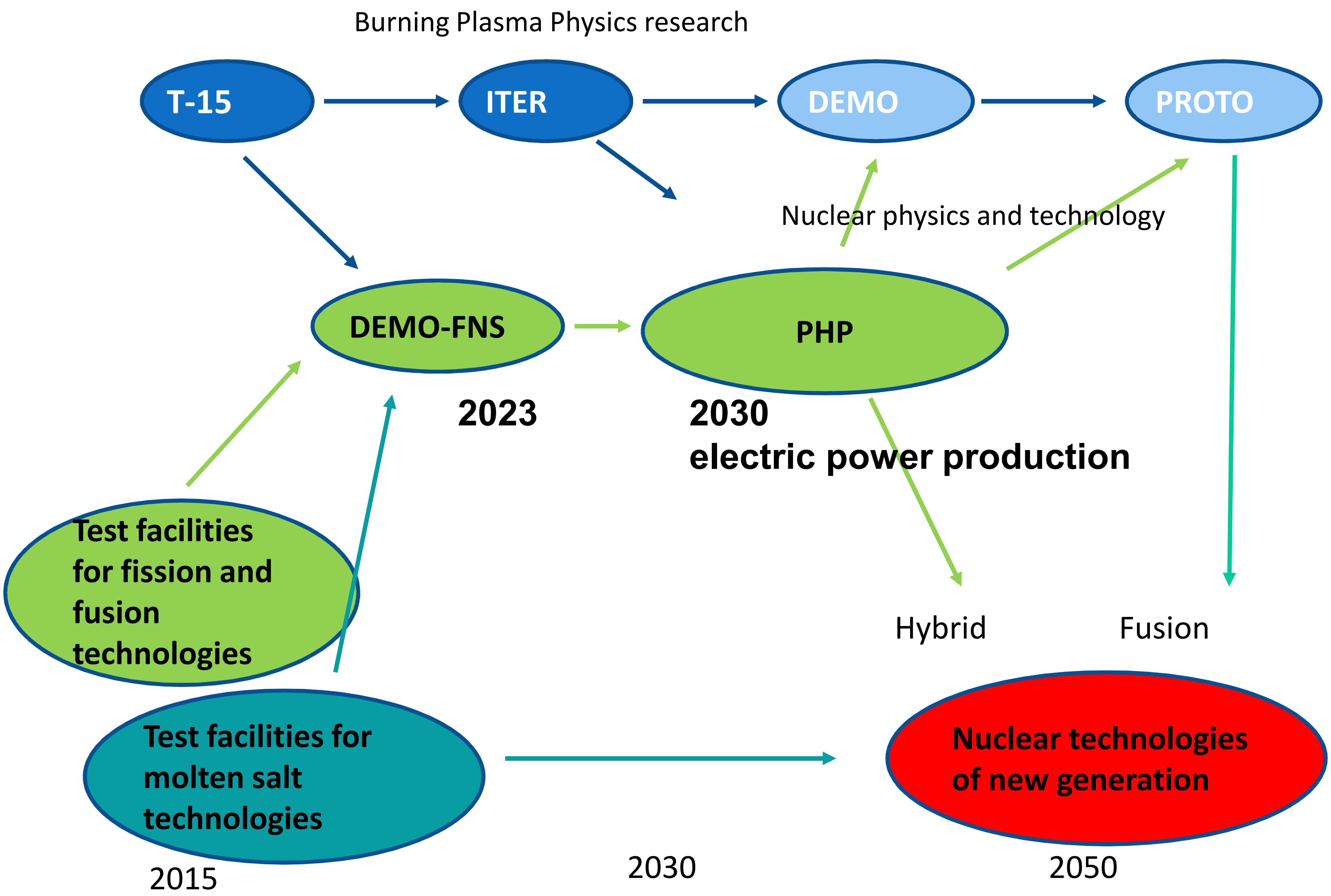
Tokomak – a source of neutrons for subcritical fission reactor

Opportunities of Hybrid Systems - more eco-friendly:

- Fission reactor – subcritical, uses external neutrons for burning. Uncontrolled reaction is excluded
- Minor Actinides transmutation
- Fuel breeding (breeding of U233 from Thorium in a molten salt blanket with suppressed fission with low radioactivity)
- Development and testing technologies and materials for DEMO project

TOKAMAKS Have a possibility to create steady state neutron sources with the yield of 10^{16}-10^{20} n/s

Strategy 2013 for Fusion-Fission development in Russia



Parameters of DEMO-FNS (conceptual) and ITER

	DEMO-FNS	ITER
Big radius R_0, m	2,75	6,2
Energy transfer from α - particles to plasma	yes	yes
Plasma configuration	DN	SN
Toroidal field at plasma axis B_{t0}, T	5	5,3
Fusion power P_{FUS}, MWt	30 - 40	500
Input power P_{AUX}, MWt	30 - 40	50 - 70
Factor of power gain, Q	~ 1	~ 10
Neutron shield, m	~ 50 см	60 – 80 см
Specific neutron load Γ_n, MWt/m^2	0,2	0,5
Neutron fluence, MWt·year/m^2	~ 2	0,3

Comparison of radiation damage effects in the first wall

Pure Fusion Reactor vs. Fusion-Fission hybrid of the same power

	Energy	Total H production	Total He production	d.p.a.
hybrid/ fusion	50	0.02	0.02	0.07

In hybrid systems requirements to materials are reduced more than order of magnitude

Perspective fusion materials

Low-activation elements: **C, Si, Ti, Fe, Cr, V, W, Mn**

1. V-(4-5)Cr-(4-8)Ti (Russia, USA, Japan, China, India) **300 – 750(800) ^{0}C**	**V-4Cr-4Ti**
2. RAFMs (Europe, Russia, US, Japan, China) **300 – 700(750) ^{0}C**	**Ek-181 (Rusfer)**
3. Austenite steels (Europe, Russia, US, Japan, China, India) **300 – 750(800?) ^{0}C**	**ChS-68**

Bochvar's Institute

4. SiC_f/SiC
(Европа, США, Япония)
до 1100 ^{0}C

- **Difficult to manufacture**
- **No experience using**

RAFM-Rusfer (EK-181, Fe-12Cr-2W-V-Ta-B-C)

Manufacturer - A.A. Bochvar High-technology Research Institute of Inorganic Materials (VNIINM), Russia
Composition – Fe-12Cr-2W-V-Ta-B-C.
Available: Almost Industrial: ingots up to 500 kg, rolled sheets, tubes,
Temperature window: (300)350 - 670(700-750) ^{0}C.

Advantages & disadvantages

+

- reduced activation
- no swelling (<100 dpa)
- ductile
- industry fabrication

-

- low heat resistance
- no high n dose tests (>100 dpa in the process),

Applications:
Fusion: TBM DEMO in ITER, DEMO-FPP (He, Pb-Li), FNS.
Fission: **FBR**s - BN-600 (2020↓), BN-800 (started 2015), BN-1200 (Na, 2020↑), BREST(Pb).

γ-phase stab., Impr. Long-term creep

Martencite formation

Strong carbide former, HT strength

γ-phase stab., Ni repl., ↓ crit. cooling rate for martecite formation

Corrosion resistance

Strong carbide former, stab. γ-phase, Mo replacement

Grain refinement, red. ↓-phase

		Ni	C	N	Si	Ti	V	Cr	Mn	Fe	Zr	Ce	Ta	W
ferritic-martensitic	Rusfer (EK-181)	0.03	0.16	0.07	0.4	0.05	0.4	**12**	0.6	**BASIS**	0.05	0.05	0.15	1.3
	EUROFER		0.1	0.015-0.045			0.15-0.25	**9.2**	0.2-0.6				0.05-0.09	1-1.2
Austenitic	ChS-68	14.0-15.5	0.05-0.08	<0.02	0.3-0.6	0.2-0.5	0.1-0.3	**15,5-17**	1.3-2.0					
	SS316 L(N)-IG	12	0.015	0.06	0.5	0.15		**17**	1.6				0,01	

RAFMs - Rusfer (EK-181, Fe-12Cr-2W-V-Ta-B-C)

Rusfer properties are under investigation in frame of Russian fission programs *(incl. tests in FBR)*

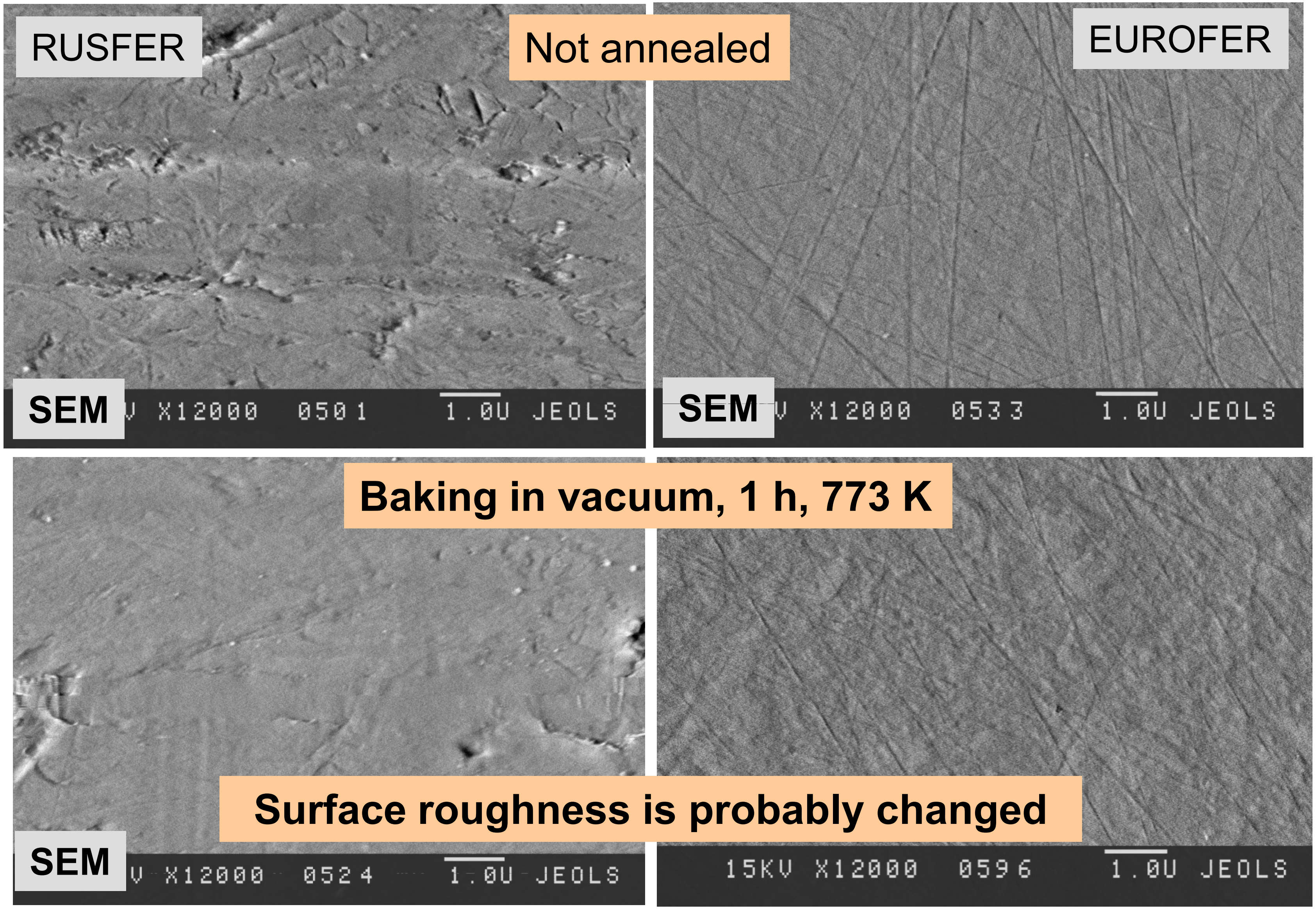
Additional investigations are necessary for fusion application:

- Hydrogen retention
- Gas and plasma-driven permeation of hydrogen
- Influence of damages on retention and permeation
- Influence of fusion neutron spectra on RAFMs properties

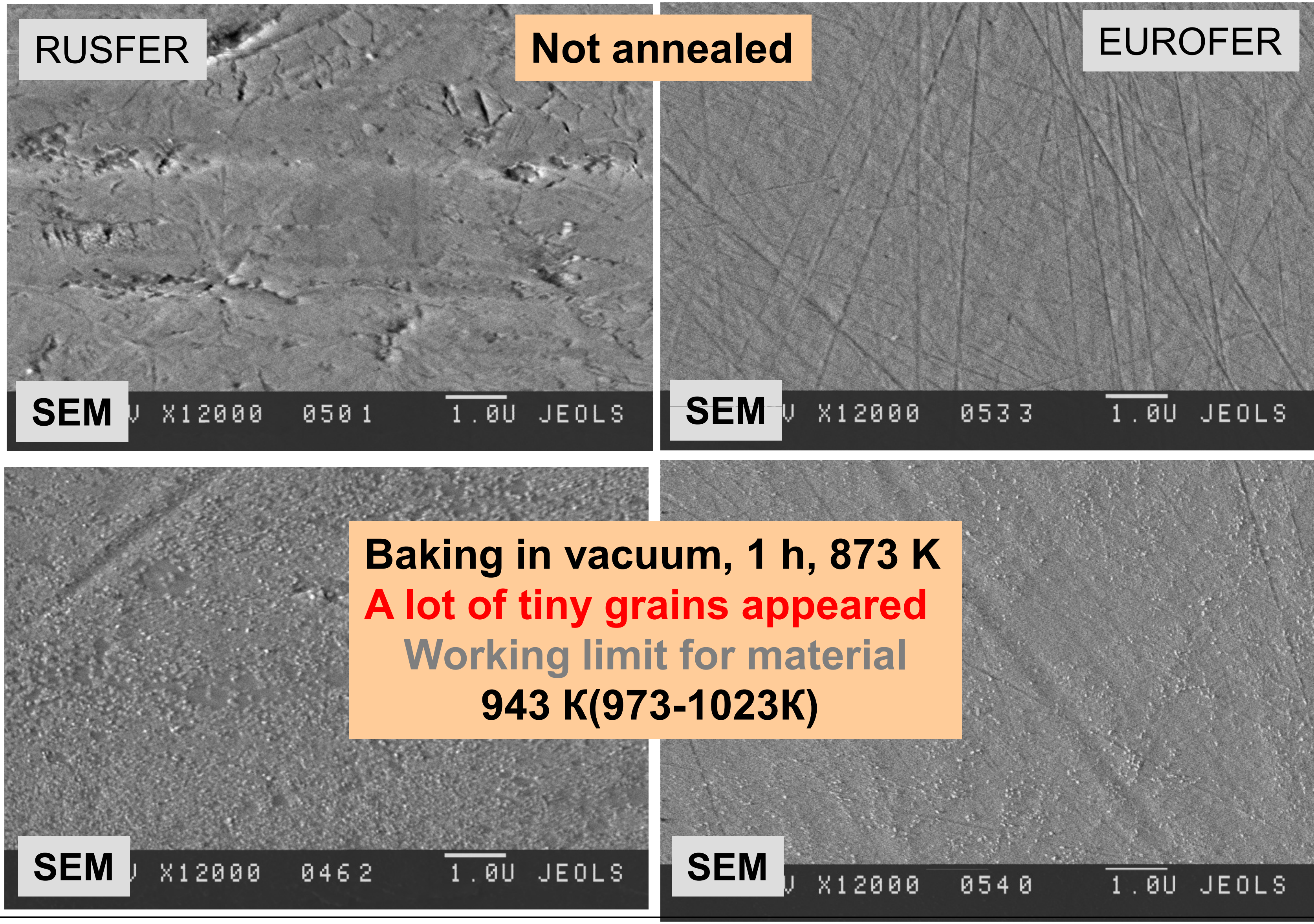
Hydrogen isotopes interaction with RUSFER – What is already done by our group

- Some microscopy
- **Deuterium retention (from gas and from plasma)**
- **Deuterium retention in damaged** (high heat fluxes, low-temperature plasma) material
- **Deuterium permeation**

Structure changes of materials at annealing



Structure changes of materials at annealing

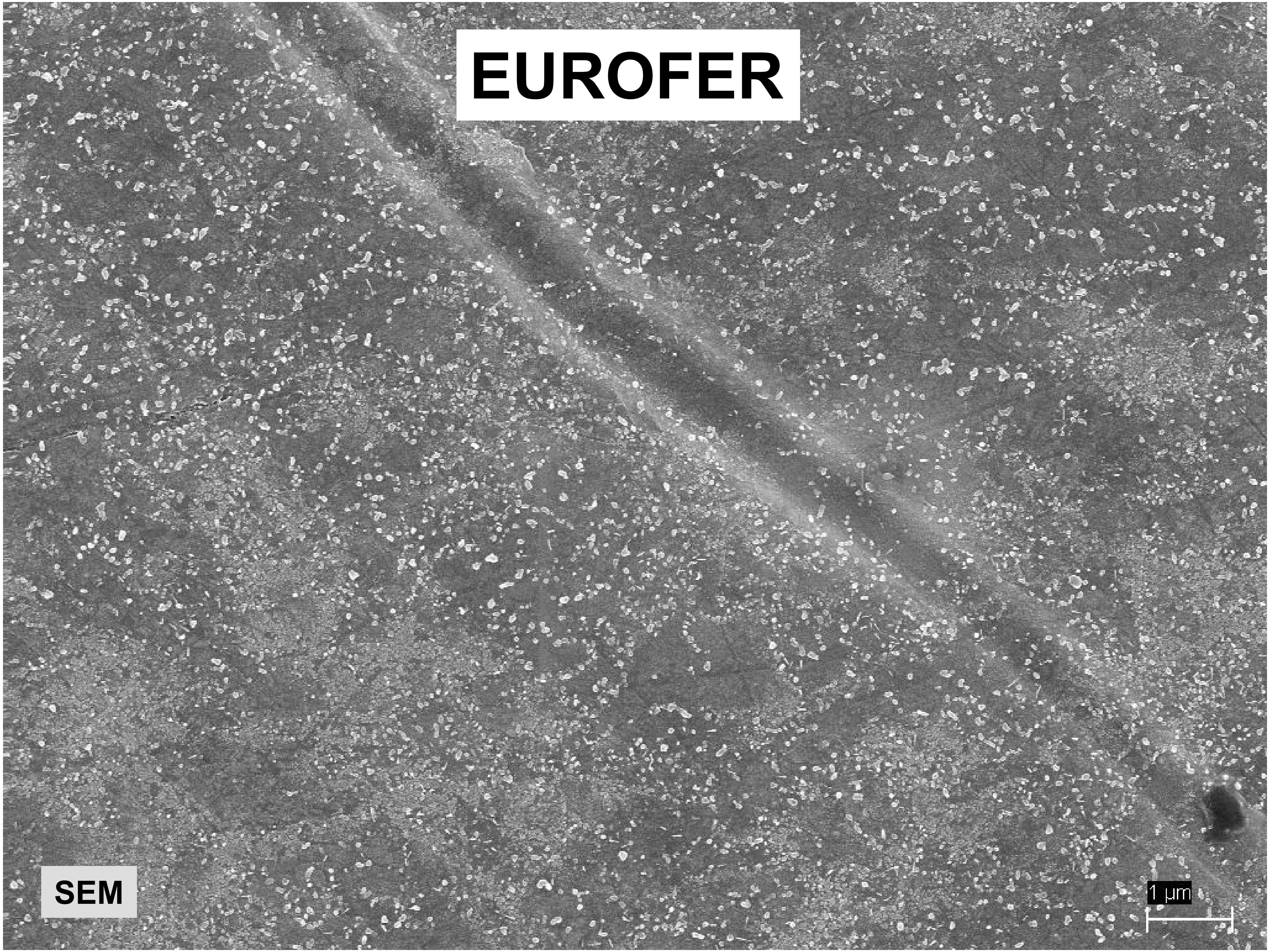


Baking in vacuum, 1 h, 873 K

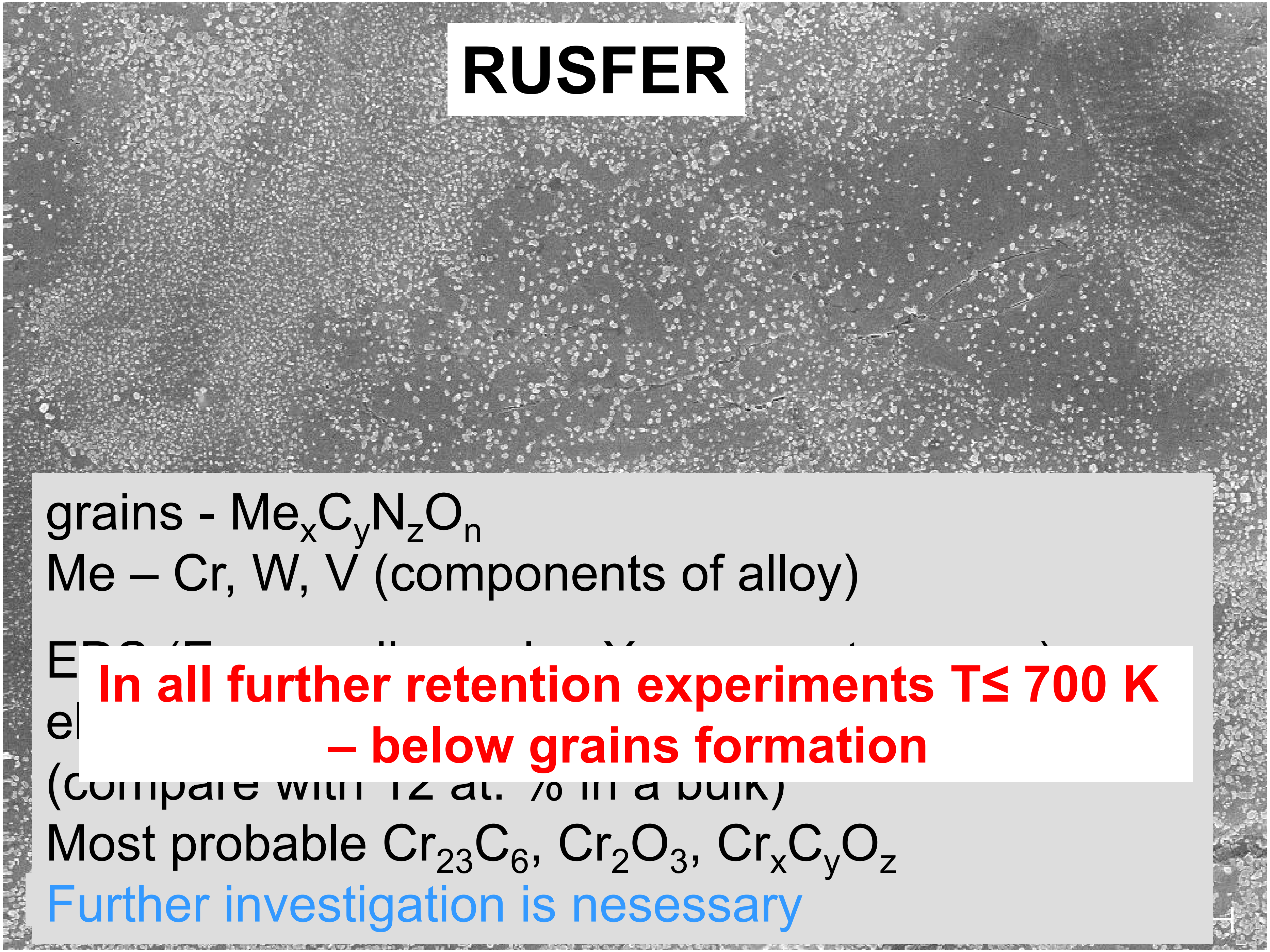
A lot of tiny grains appeared

Working limit for material

943 K(973-1023K)


EUROFER
SEM
1 µm

RUSFER



grains - $Me_xC_yN_zO_n$
Me – Cr, W, V (components of alloy)

E

el

In all further retention experiments T≤ 700 K – below grains formation

(compare with 12 at. % in a bulk)
Most probable $Cr_{23}C_6$, Cr_2O_3, $Cr_xC_yO_z$
Further investigation is nesessary

Deuterium retention in RUSFER from gas

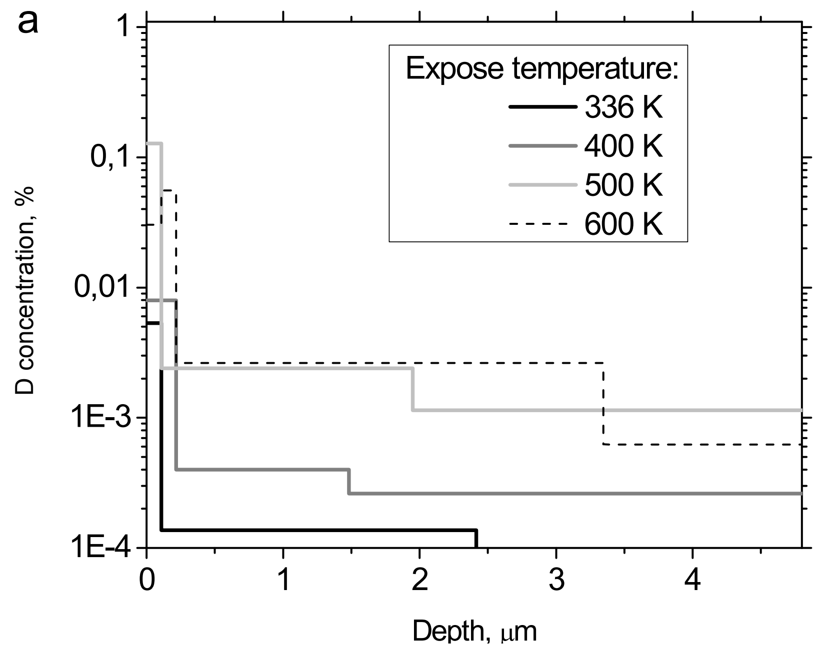
Gas loading:

D_2,

10 or 10^4 Pa

1-16 h

RT-600 K



Main peculiarities:

- Deuterium retention is small at T<450 K and **increases with increasing temperature**;
- Concentration of D in RAFMs bulk is relatively low (**10^{-3}-10^{-2} at%**)
- Deuterium retention is much higher in the very near-surface layer (~0.2 mkm);
-The dependence of D retention on gas pressure is very weak (at T=693 K the difference between the amount of deuterium retained at P=10 Pa and at P=10^4 Pa is only by about a factor of 2).

Golubeva et. Al., J. Nucl. Mater. 438(2013) S983-S987

Deuterium retention in RUSFER at plasma irradiation

Plasma-irradiation

Temperature	RT – 700 K
Gas pressure (D_2)	$5{\cdot}10^{-4}$ mbar
Plasma composition	D_3^+ - 70%, D_2^+ - 20%, D^+ - 10%
Accelerating potential:	- 300 V
Plasma flux	$6{\times}10^{19}$ at/s×m^2
Plasma fluence	$10^{23} \div 10^{25}$ D/m^2

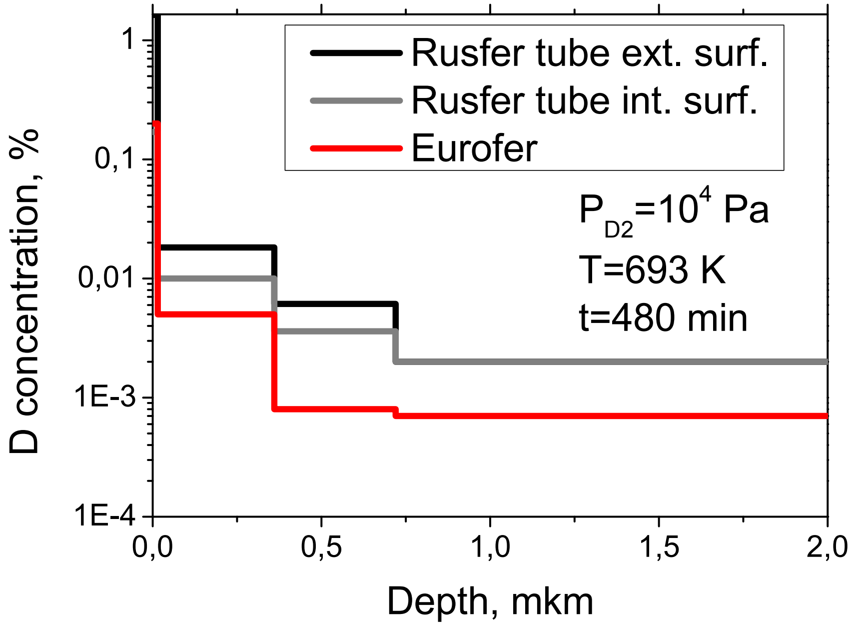
Plasma-irradiation

- Constant cleaning of surface layer
- Elevated concentrations in near-surface layer
- Defects creation in near-surface layer

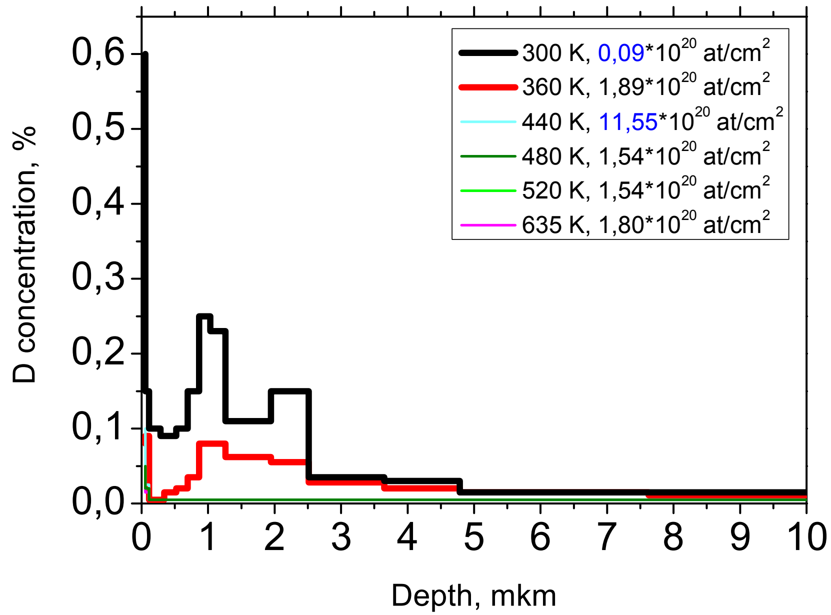
Golubeva et. Al., J. Nucl. Mater. 438(2013) S983-S987

NRA (Depth profiles), IPP Garching

Gas exposure



Plasma irradiation



Much higher concentrations under surface can be achieved as compared with gas exposure

Deuterium retention in damaged RUSFER

1. Damage:

- 20 MeV W^{6+} ions (0.94 dpa) – modeling of neutrons damage – IPP
- Plasma $5{\cdot}10^{21}$ $D/s{\cdot}m^2$, 10^{25} D/m^2 - LENTA – Kurchatov institute
- Heat load (10 pulses of 0.5 MJ/m^2, 0.5ms) - QSPA TRINITI

2. Gas loading

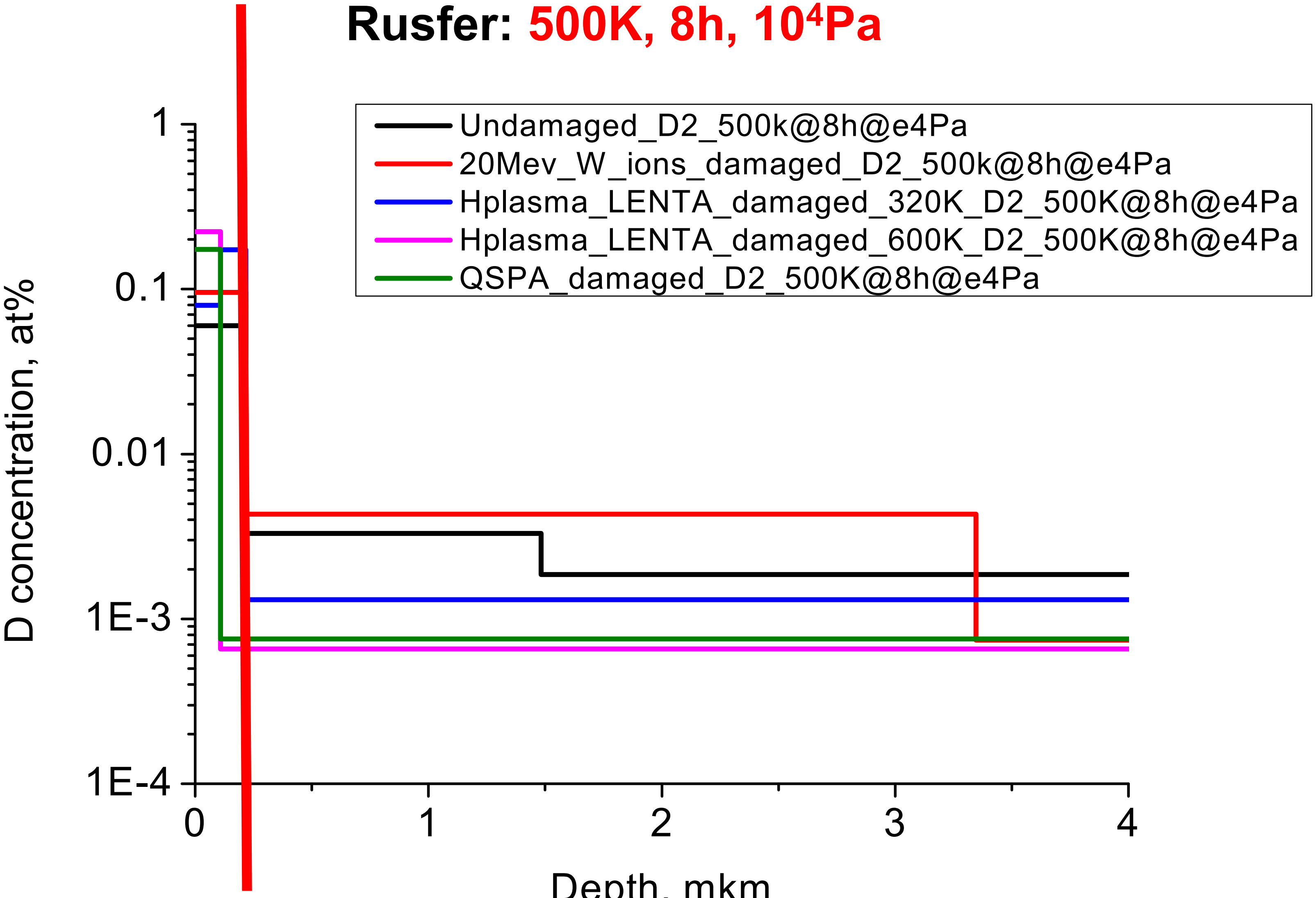
Atlan (10^4 Pa, 8 h, RT-600 K) – Kurchatov institute

3. Retention analysis

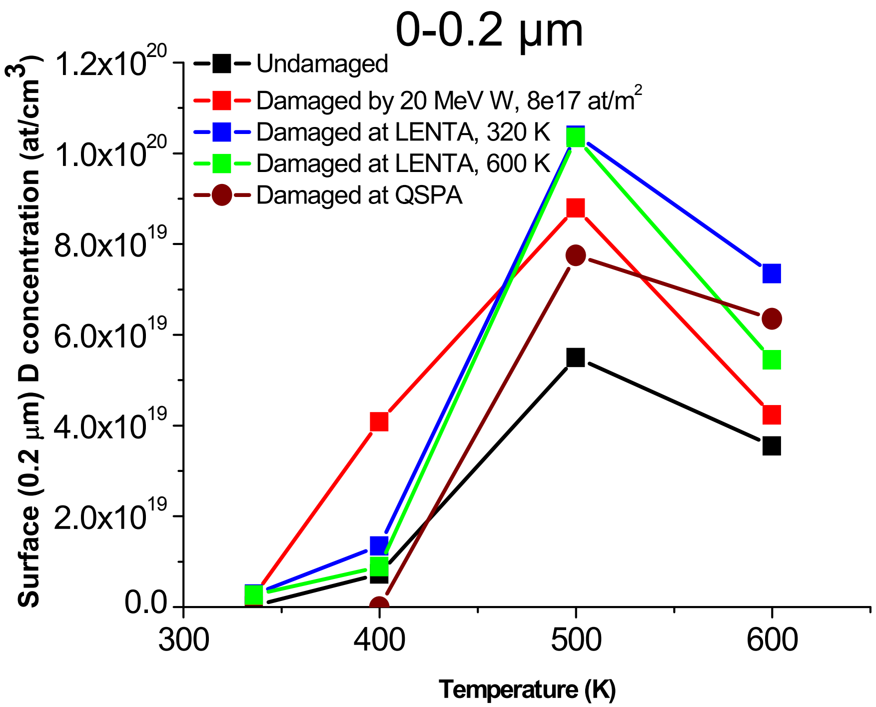
NRA $D(^3He, p)\alpha$ – IPP

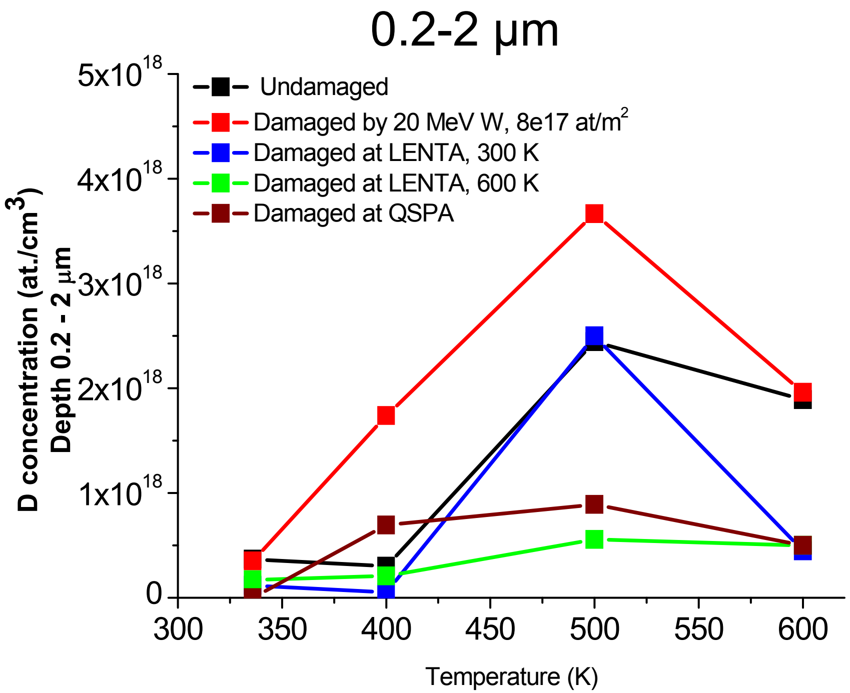
A.V. Spitsyn et. al., J. Nucl. Mater. 455 (2014) 561-567

Deuterium retention in damaged RUSFER



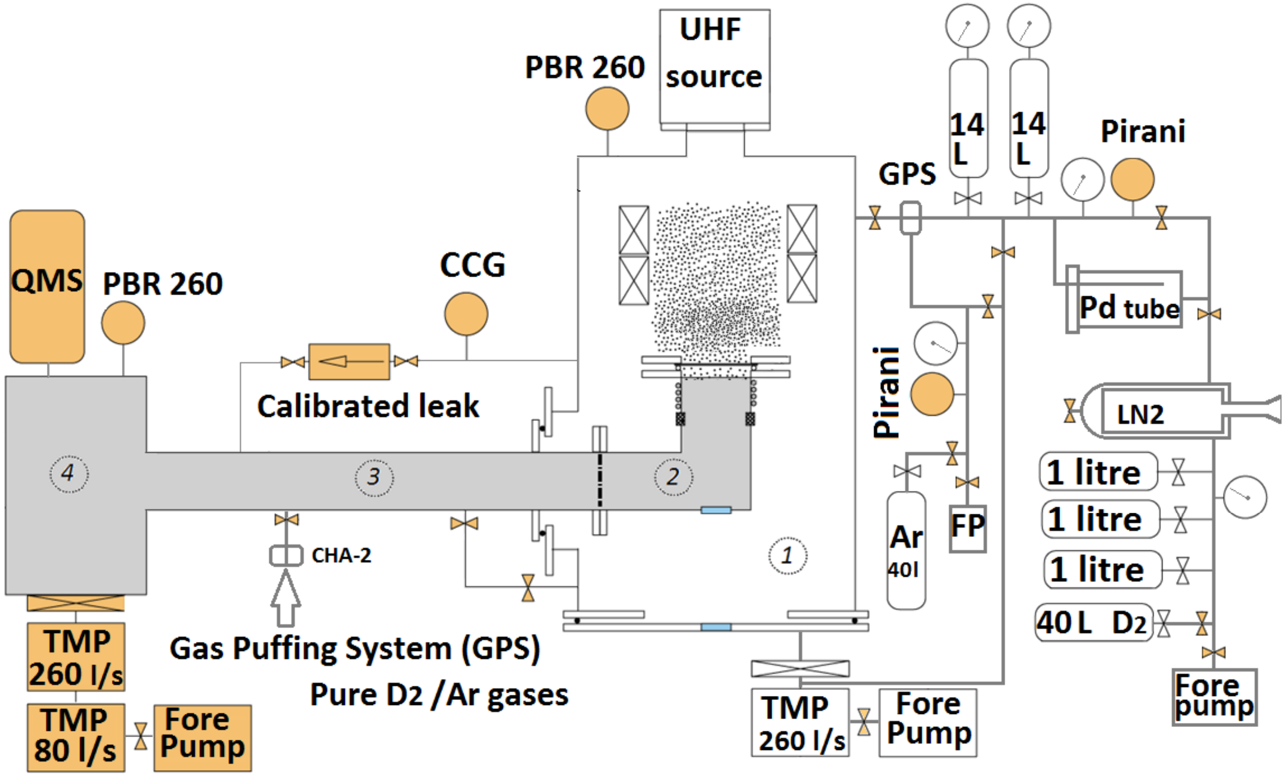
Deuterium retention in damaged RUSFER





- The maximum D retention in all samples were observed after expose to D_2 at 500 K
- D mainly trapped in near-surface layer (~0.2 mkm)
- Concentrations of deuterium trapped in damaged samples are close to those in undamaged samples

Deuterium permeation, PIM, Kurchatov institute



Membrane temperature: RT – 1000 K

Gas pressure (D_2 or Ar) **$5 \cdot 10^{-5} \div 1 \cdot 10^{-3}$ mbar**

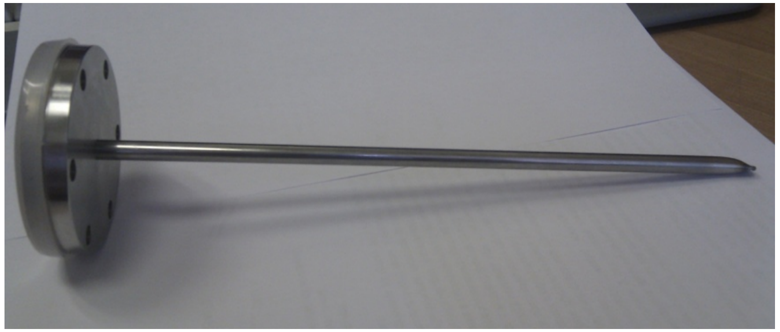
Plasma composition: ions: 71% (D_3^+:D_2^+:D^+ → 7:2:1) neutrals: 29% D_1^0

Ions accelerating potential: up to -300 V

Plasma density: up to $6 \cdot 10^{10}$ cm^{-3}

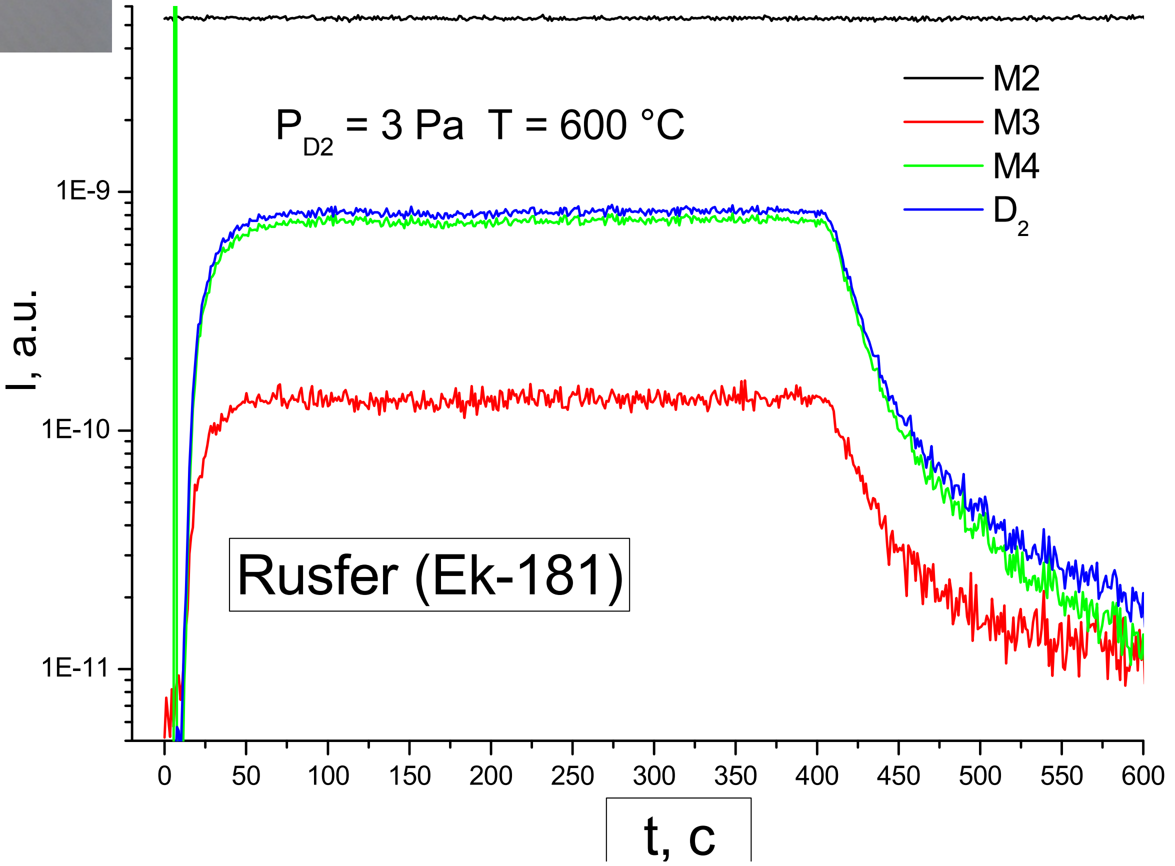
Plasma current on sample: up to 20 $A \cdot m^{-2}$

Gas-driven permeation

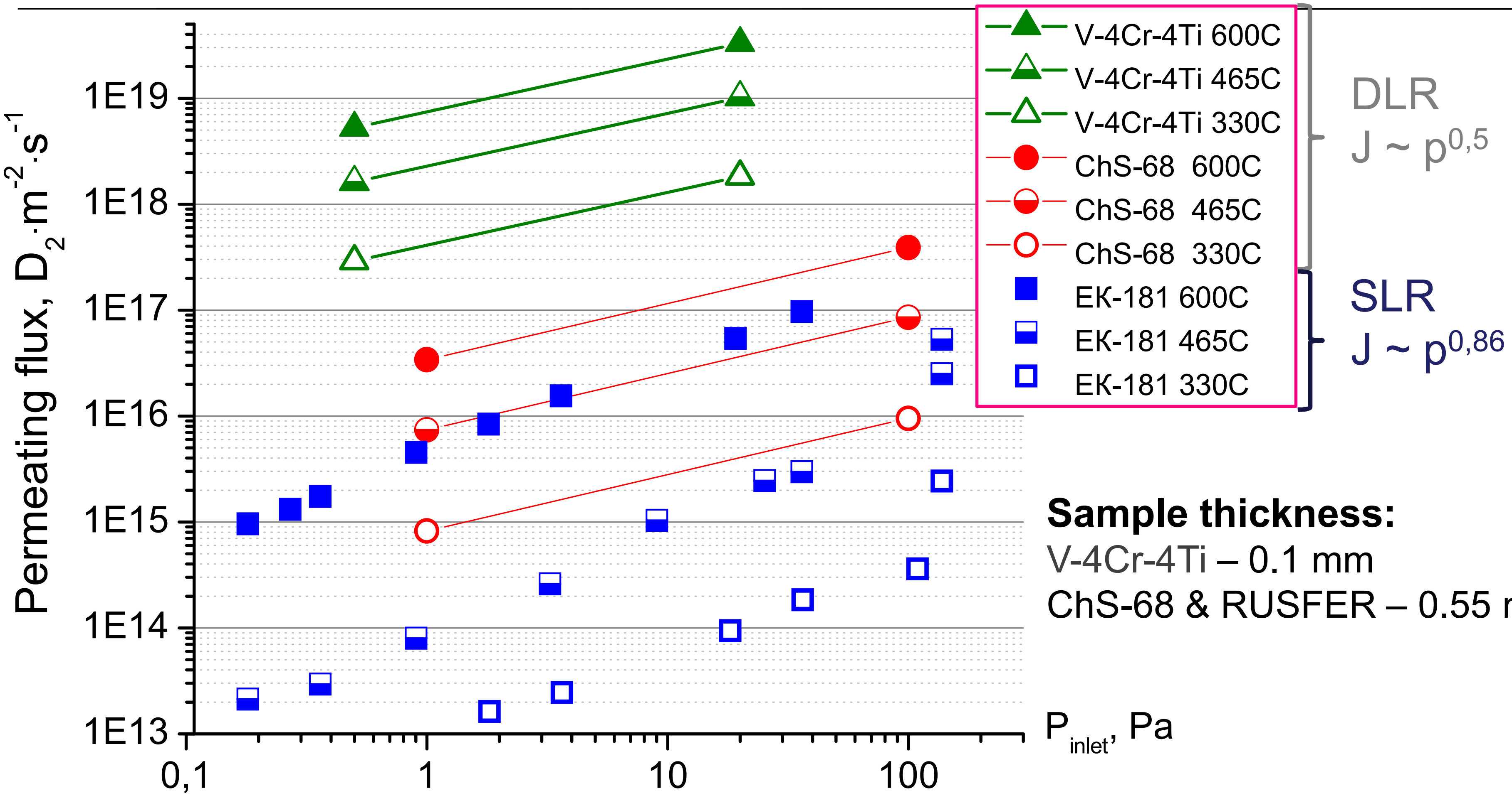


- **Rusfer EK-181**
- **ChS-68**

Tubes
L=250 mm
d1= 5,85 mm
d2 = 6.95mm



Deuterium permeation



Sample thickness:
V-4Cr-4Ti – 0.1 mm
ChS-68 & RUSFER – 0.55 mm

PhD of D.I. Cherkez, planned for publishing

Deuterium permeation

Permeation of ChS-68:

- **DLR** at p>1 Па, **P= 1.24·10^{-7}·e $^{(-60.2 [kJ/mol] / RT)}$** [mol/(m·sec·Pa^0.5)]
- Diffusivity: **D = 3,7·10-6·exp {-59 [kJ/mol] / RT} [m²/sec]**
- correlates well with literature data for austenitic steels, including SS316

Permeation of EK-181:

- **J ~ p^{0,86} - transition permeation regime**
- Estimated Permeation constant for T = 600C

P = D · S = JD/p^{0,5} = 3.3e-12 ± 20% [mol/(m·sec·Pa^0.5)] (*under estimation*)

smaller than for an Eurofer about factor of 30*

=> **Needs some additional experiments for DLR obtaining.**

*(*D. Levchuk et al. //Journal of Nuclear Materials 328 (2004) 103–106)*

	S, m²	**d, mm**	**p, Pa**
DEMO	1200	10	10
FNS	100	3	

Tritium leakages for **DEMO & FNS** [grams/year]:

DEMO	T=330C	T=465C	T=600C
V-4Ti-4Cr	2431	13507	44162
ChS-68	29	261	1189
EK-181	1	10	311

FNS	T=330C	T=465C	T=600C
V-4Ti-4Cr	675	3752	12267
ChS-68	8	72	330
EK-181	0,1	3	86

Current project plans

Motivation:

Neutron irradiation ⟹

- elements transmutation
- He and H formation
- helium bubbles formation
- defects

⟹ Influence H interaction with material

No fusion neutron sources are available

Possible experimental methods of modelling of n-irradiation influence:

- **Irradiation in nuclear reactors** (another spectra, activated samples $\Rightarrow$ hot cells)
- **Irradiation by heavy ions** (cascades instead of point defects, few μm damaged layer, no uniform defect distribution). **IPP**
- **He ions implantation** (few μm damaged layer, no uniform defect distribution)
- **Irradiation by high energy electrons for defects production** (small cross-section, few mm damaged layer, uniform defect distribution)
- **He formation after tritium saturation** (time-demanding, low He concentration, uniform distributions)

The aim of the project:

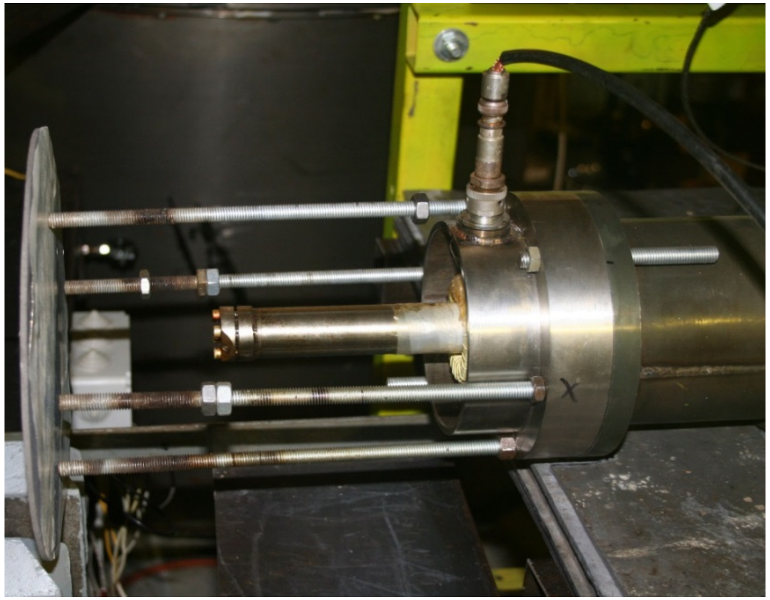
Investigation of influence of defects

(MeV electrons irradiation, radiogenic He)

on deuterium retention in RAFM

MeV electrons irradiation: ECHO, Kurchatov institute

Pulse current	100 mA
Pulse duration	5 μs
Frequency	100 Hz
Beam spot	ø 5-8 mm
Current density	**0.2-0.5 A/cm²**
Sample temperature	RT ÷ 50 ^{0}C



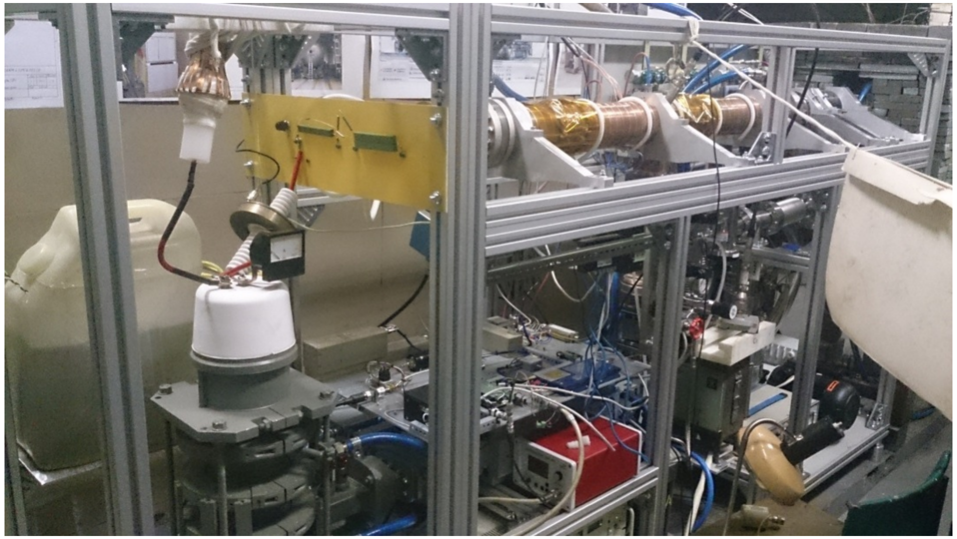
ECHO Electron accelerator

3 MeV

- **Easily accessible**
- **Broken at the moment**

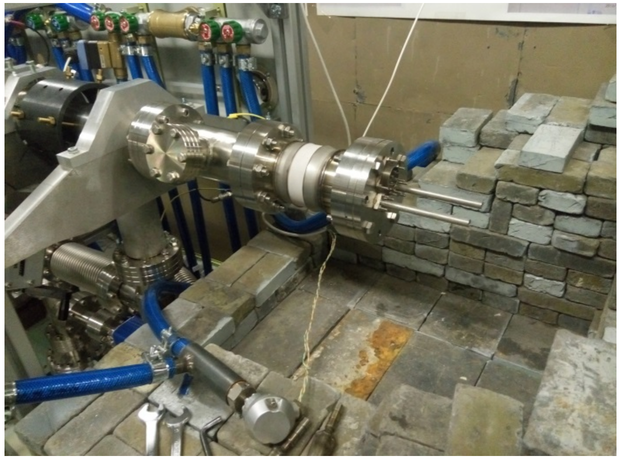
MeV electrons irradiation: Electron accelerator MSU

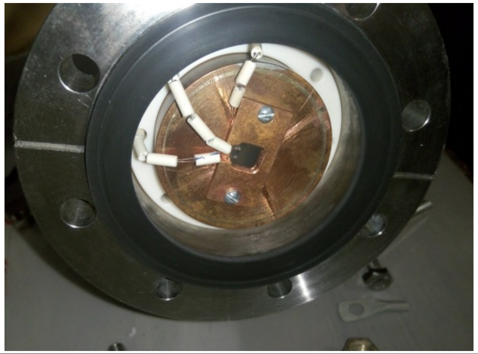
Pulse current	400 mA
Pulse duration	7 μs
Frequency	10-100 Hz
Beam spot	ø 10-50 mm
Current density	**0.02-0.5 A/cm^2**
Sample temperature	50 ÷ 150 ^{0}C

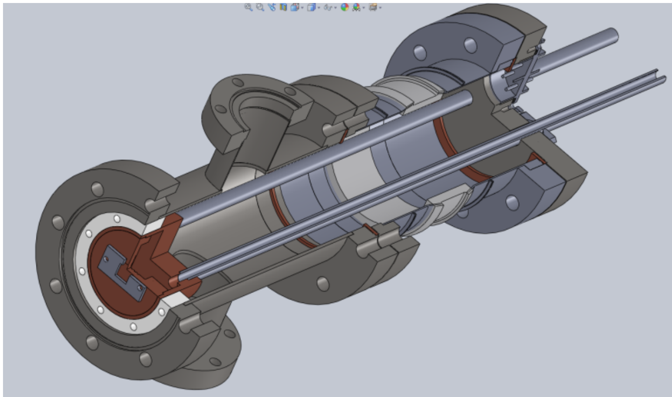


MSU Electron accelerator

10 MeV



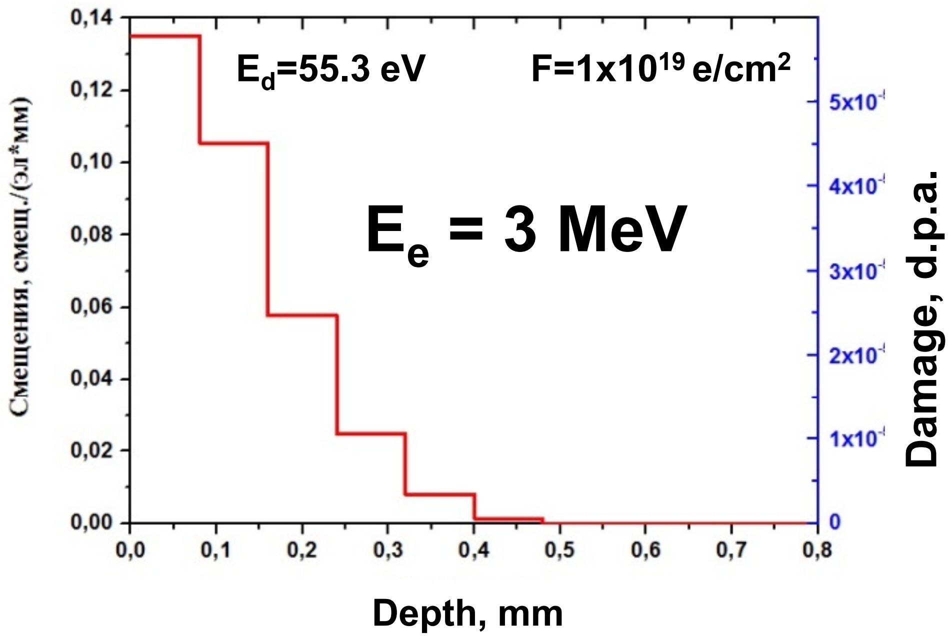




Developed sample holder

Influence of e-irradiation on H retention in W

Damage of W by 3 MeV electrons at 300 – 320 K

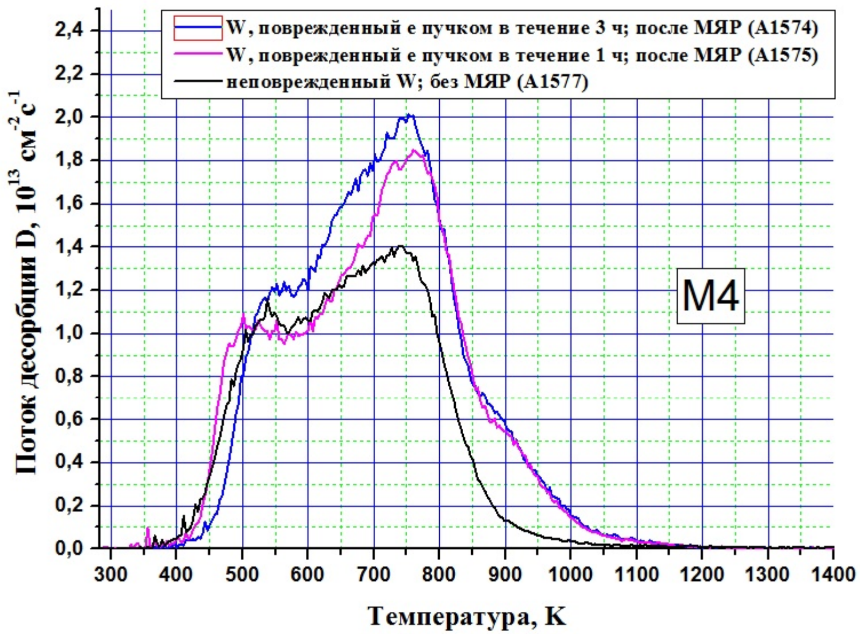


Damage depth-destribution in W after e-irradiation (3.5 MeV, 2,7·10^{18} e/м^2, 3 hours. E_d=55.3 eV

Calculations of M. Zibrov in ElectronDamage code, 2014

Not cascades but only point defects and small clusters are created

D retention in damaged W



TDS of W irradiated 10 MeV electrons (not irradiated, 1 hour, 3 hours of irradiation)

Instrument for identification Frenkel pair defects

Methods of creating He in material bulk

Methods of creating He in material bulk:

• n-irradiation (nuclear reactoins – radioactive samples)
• He implantation (not uniform distribution)
• Radioactive decay of tritium dissolved in material ("tritium trick")

$${}_1T^3 \rightarrow {}_2He^3 + e^- + \overline{\nu}$$

Tritium trick is a way co saturate material bulk with He:
- uniform depth-distribution
- without creating defects of crystalline structure (like irradiation does)

Experimental procedure:

1. Tritization of samples (exposition in tritium)
2. Holding samples with T for ^{3}He generation
3. Detritization of samples
4. Measurement of amounts of remaining T and produced ^{3}He

“Tritium trick“

Planned to investigate He production in Eurofer and Rusfer RAFMs

Work with T is planned to perform in A.A. Bochvar’s institute (have license). Special volume (120 cm^3) is under construction for filling with high pressure tritium.

Two expositions are planned:

1. 500^0C, 50 atmospheres, several hours
2. 250^0C, ~20 atmospheres, $\leq$ 10 hours

Samples holding with tritium – 3, 6, 9 months

Detritisation – heating (700^0C), exposition in H for isotope exchange

Controlling amount of T - radioluminescence (destructive method)

Controlling amount of ^{3}He – calculations, mass-spectrometry at melting

Remaining radioactivity of sample 15x12x1 mm **< 10^9 Bq** (can be handled without special protection)

No warranty that remaining radioactivity would be sufficient for transfer to other institution, but in any case result on He generation would be interesting itself.

Plans in frames of Project:

Year 1:

- Investigate the influence of a dense low-energy plasma irradiation on the morphology of RAFM steel using the LENTA facility in the Kurchatov nstitute
- Investigate the influence of pulsed heat loads on the morphology of RAFM steel

Year 2:

- Prepare and investigate RAFM samples with introduced radiogenic helium
- RAFM samples damaged using high-energy electrons
- RAFM samples damaged using high-energy heavy ions

Year 3:

Investigate the influence of introduced radiogenic helium and of damage by high-energy electrons and ions on deuterium retention in RAFM steel

What is required:

1. **Experimental modeling of different defects on hydrogen retention and permeation throw RAFMs**

2. **Influence of neutron effects on mechanical properties and thermal conductivity of RAFMs**

Thank you for your attention!