Lead-Bismuth Eutectic Alloy as a Coolant: Benefits and Challenges

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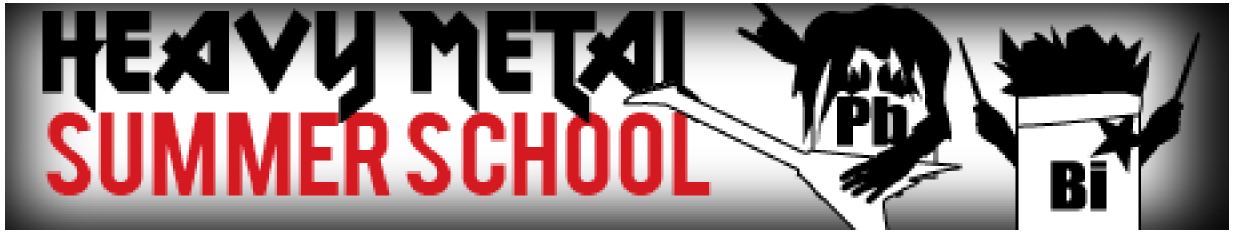
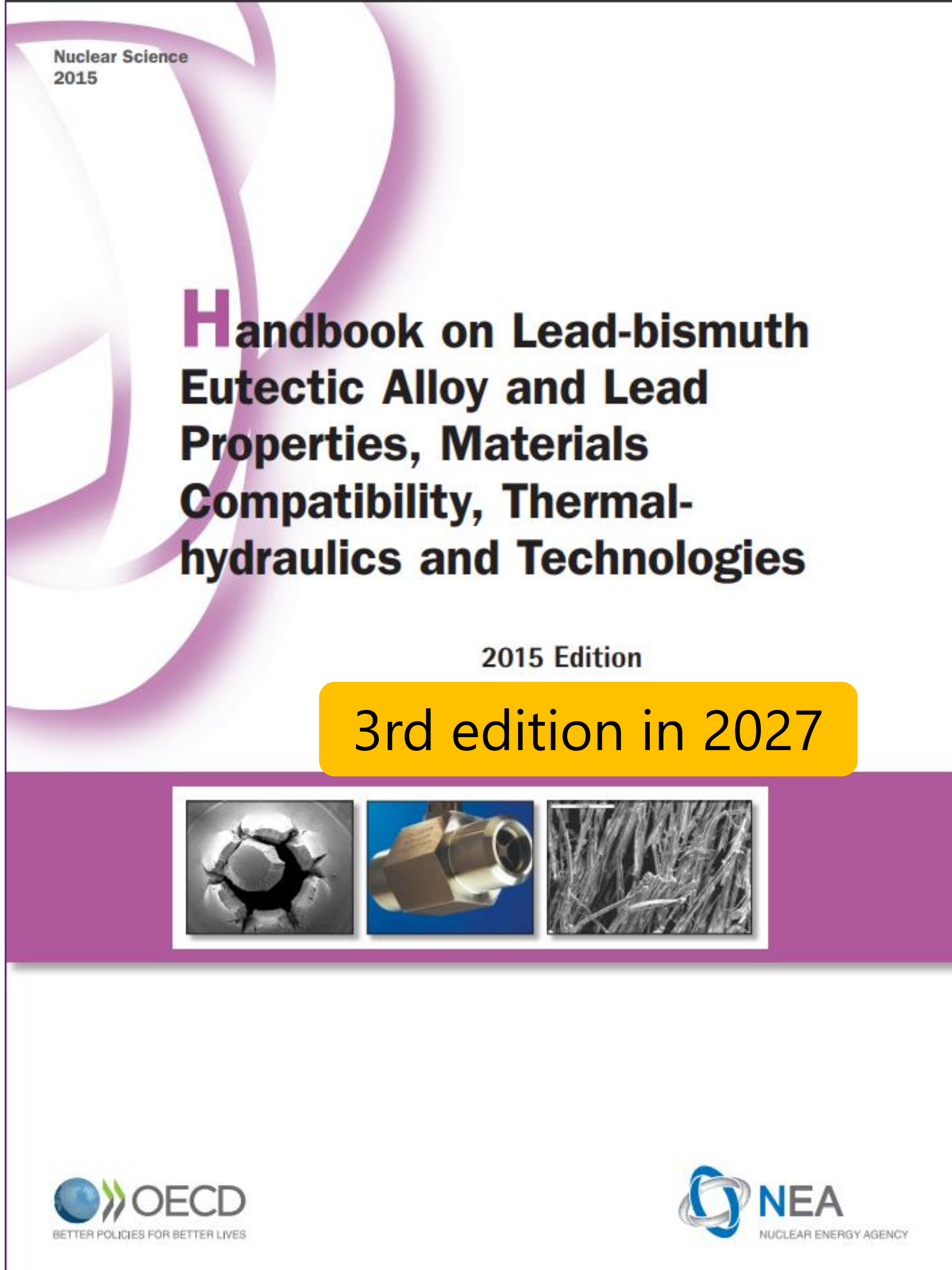
IAEA National training course on Heavy Liquid Metal Cooled Fast Reactors

Pitesti, Romania

Scope of this lecture

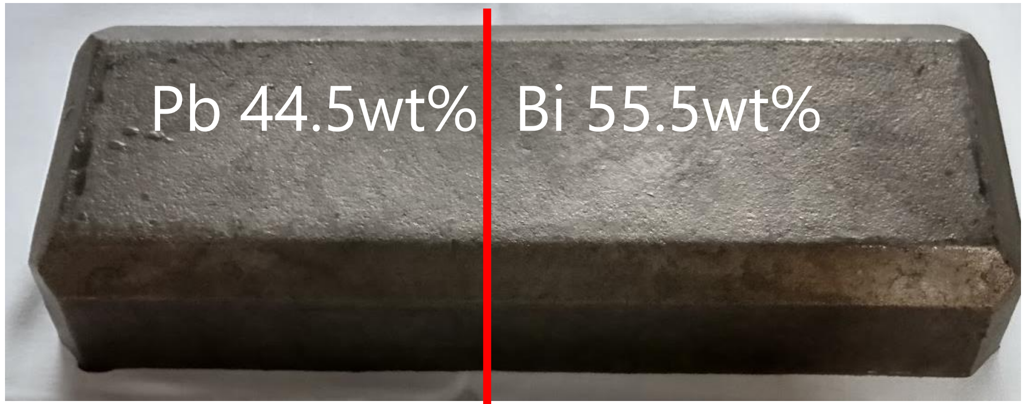
- Only 1 hour, cannot cover everything
- Focus on specific features of LBE vs. pure lead, mainly **coolant chemistry**
- See also: "*Fundamentals of LFR technology and Accelerator Driven Systems (ADS)*"

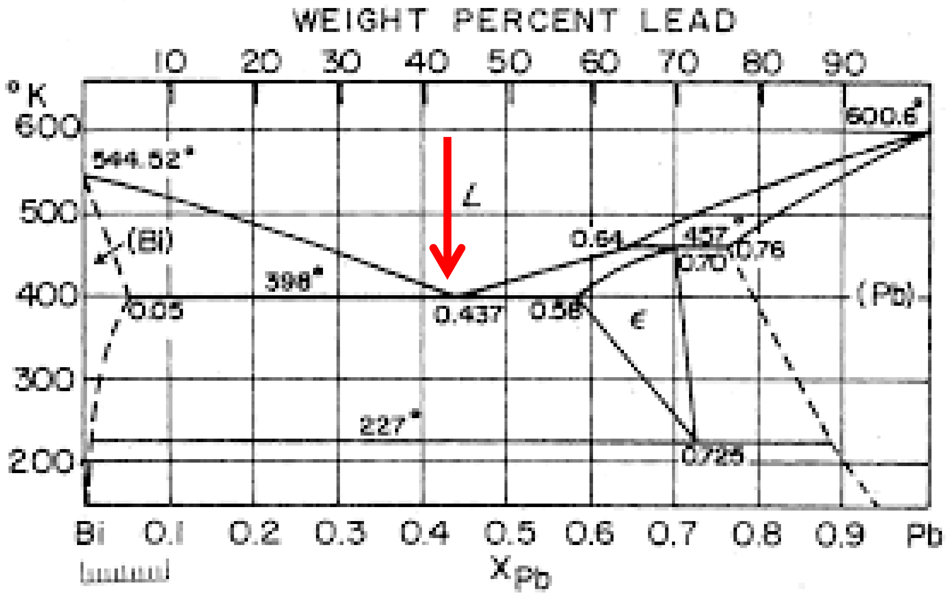


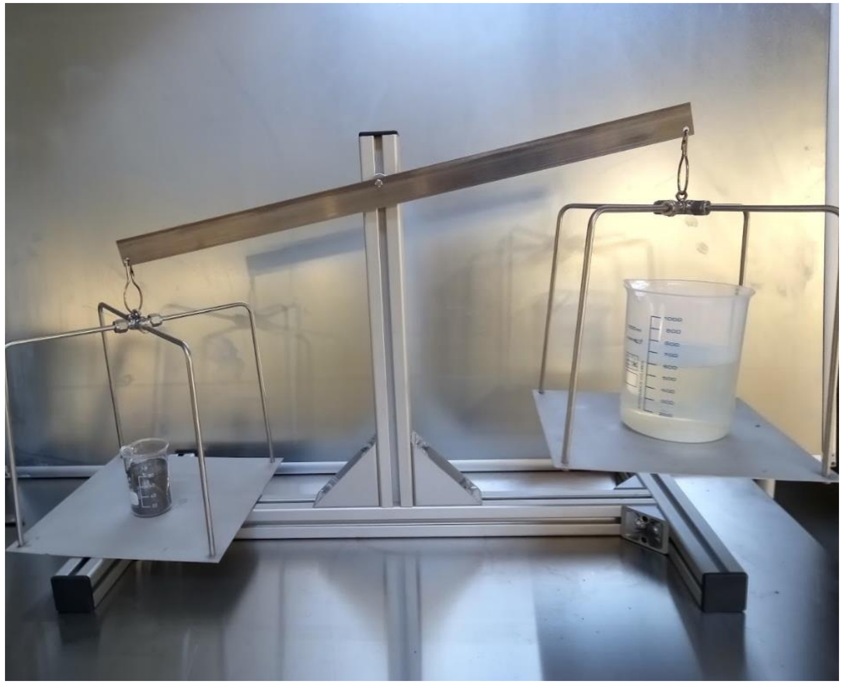


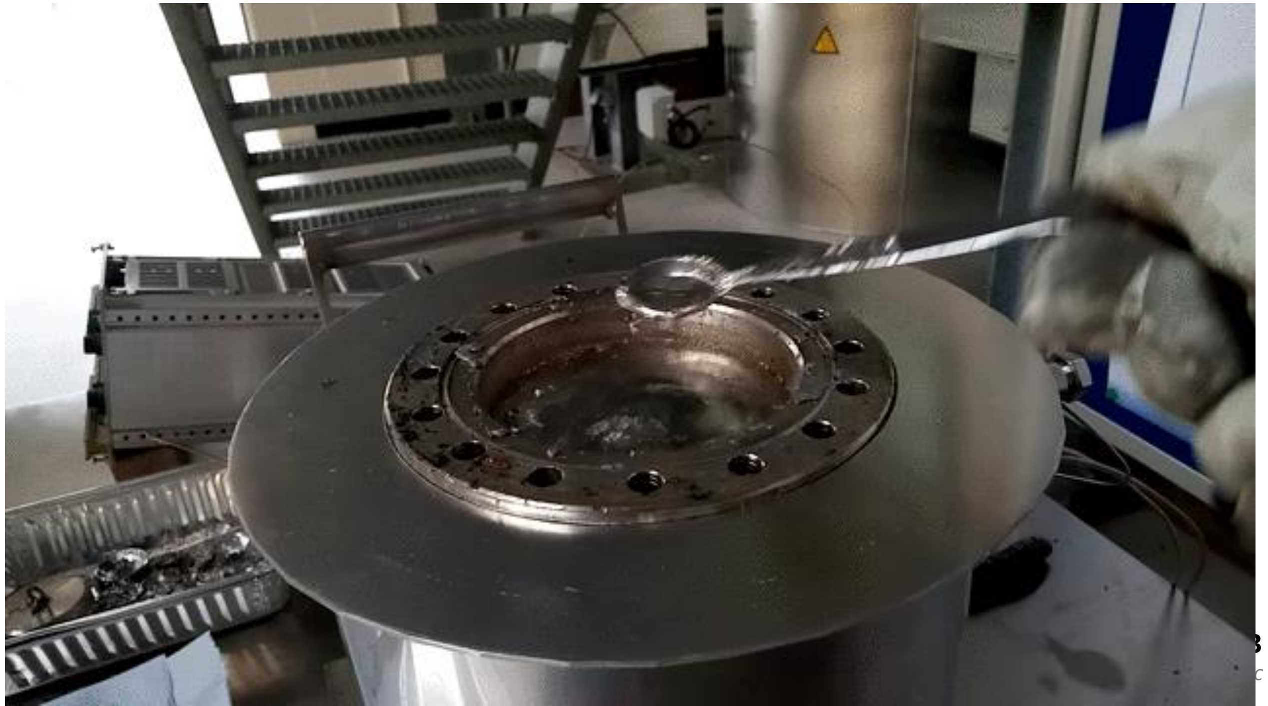
3rd edition in 2027

LBE = Lead Bismuth Eutectic









Pure lead or lead-bismuth eutectic (LBE)?

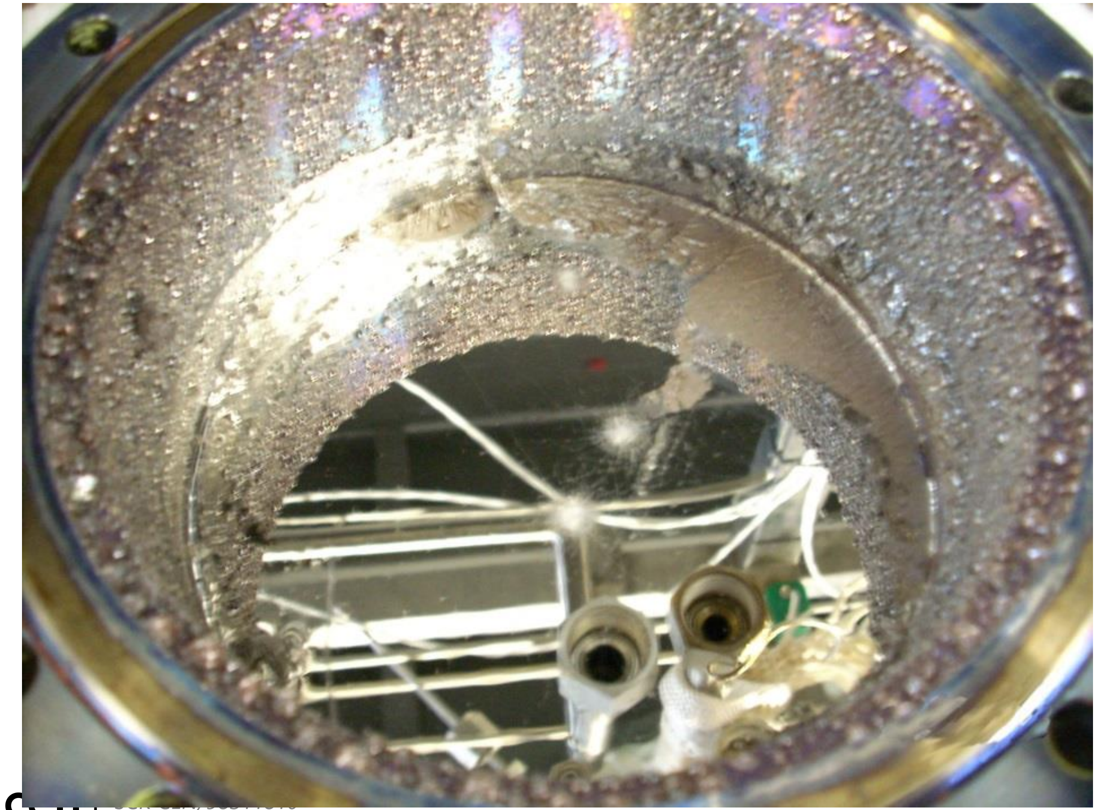
Price ~x10

Property	Units	LBE	Lead	Sodium	Water
Melting temperature	°C	125	327	98	0
Boiling temperature	°C	1654	1748	882	100
Reference temperature	°C	200	400	500	25
Density	kg/m^3	10453	10579	832	997
Kinematic viscosity	m^2/s	2.33 E-7	2.10 E-7	2.88 E-7	8.93 E-7
Thermal conductivity	$W/(m \cdot K)$	10.4	16.6	67.7	0.6
Specific heat capacity	$J/(kg \cdot K)$	146.9	146.7	1264	4182
Prandtl number	-	0.0343	0.0197	0.0047	6.1366
Surface tension	N/m	411 E-3	453 E-3	156 E-3	72 E-3
Speed of sound	m/s	1755	1787	2316	1498
		HLM Handbook (2015)		*SCK CEN-BLG-1069*	*IAPWS-97*

Primary Coolant must be *clean* at all times

'Clean' LBE

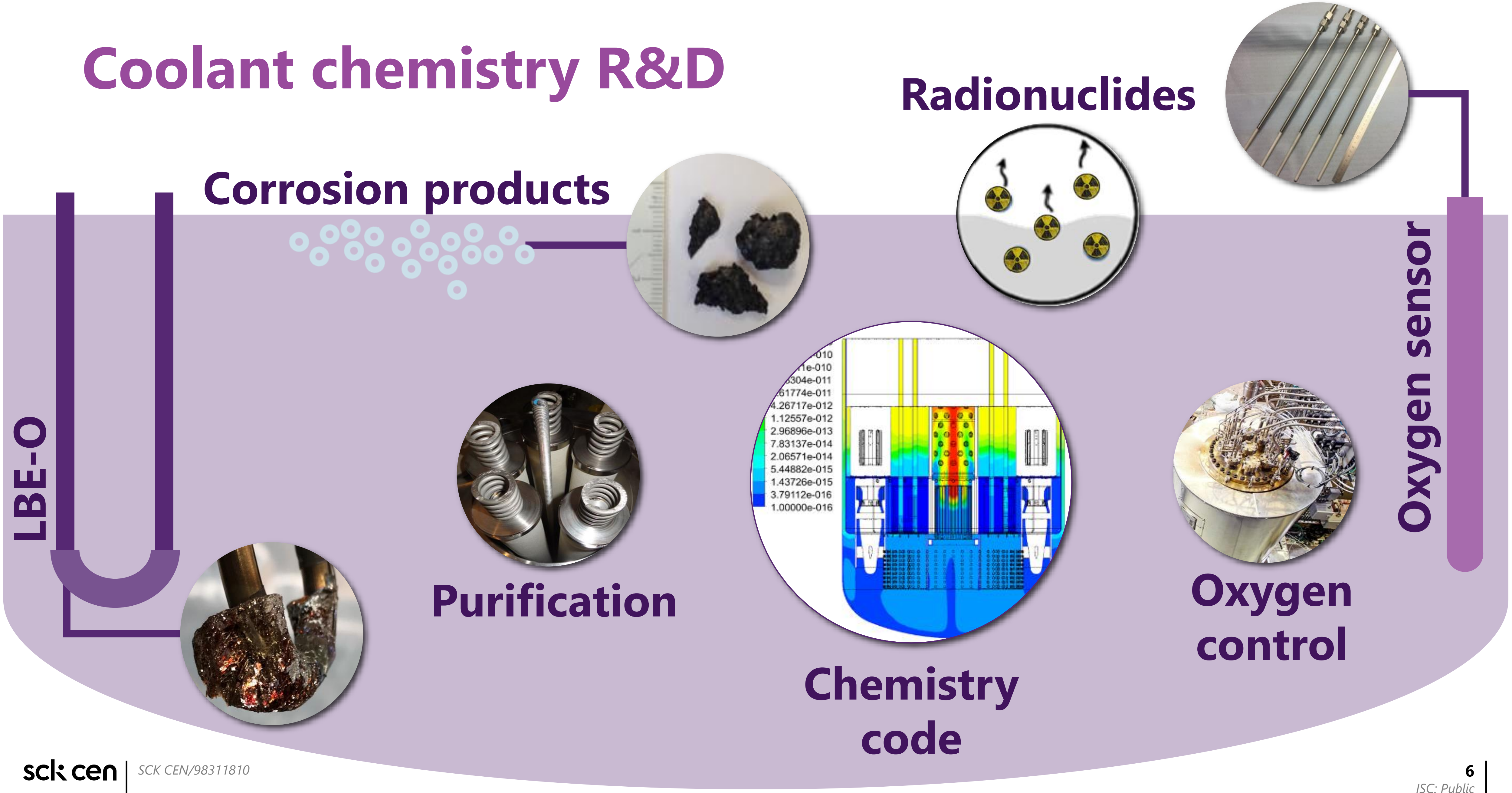
shiny liquid, like a mirror



'Dirty' LBE

Solid PbO precipitates floating

Coolant chemistry R&D

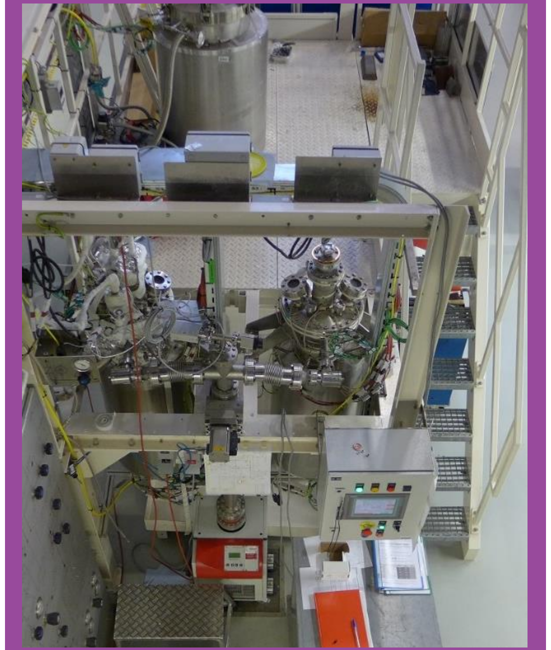


Coolant chemistry experimental facilities

MEXICO

Integrated LBE Chemistry Research Loop

HELIOS



LBE-Gas Interaction

POVACS

Polonium Evaporation and Sticking in Vacuum

CHECKMATE

Small-scale LBE Chemistry Study

Coolant chemistry experimental facilities

SPLAT

LBE Aerosols

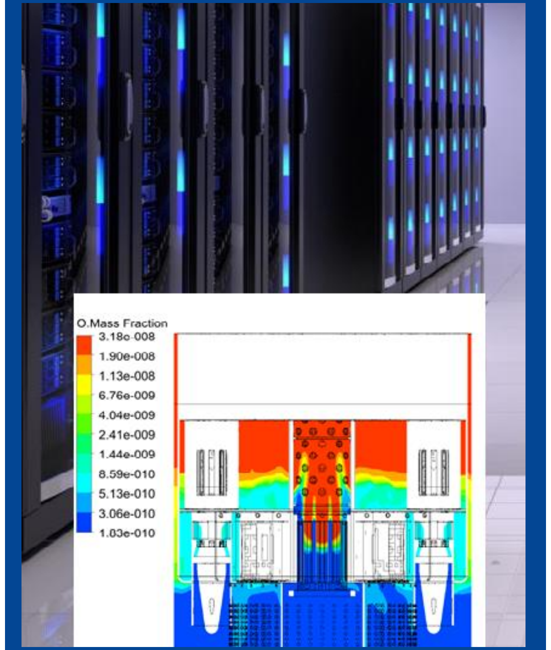
LBEAM

Radionuclides Evaporation from LBE

Polonium Transpiration

Polonium Evaporation from LBE

GEM-CFD



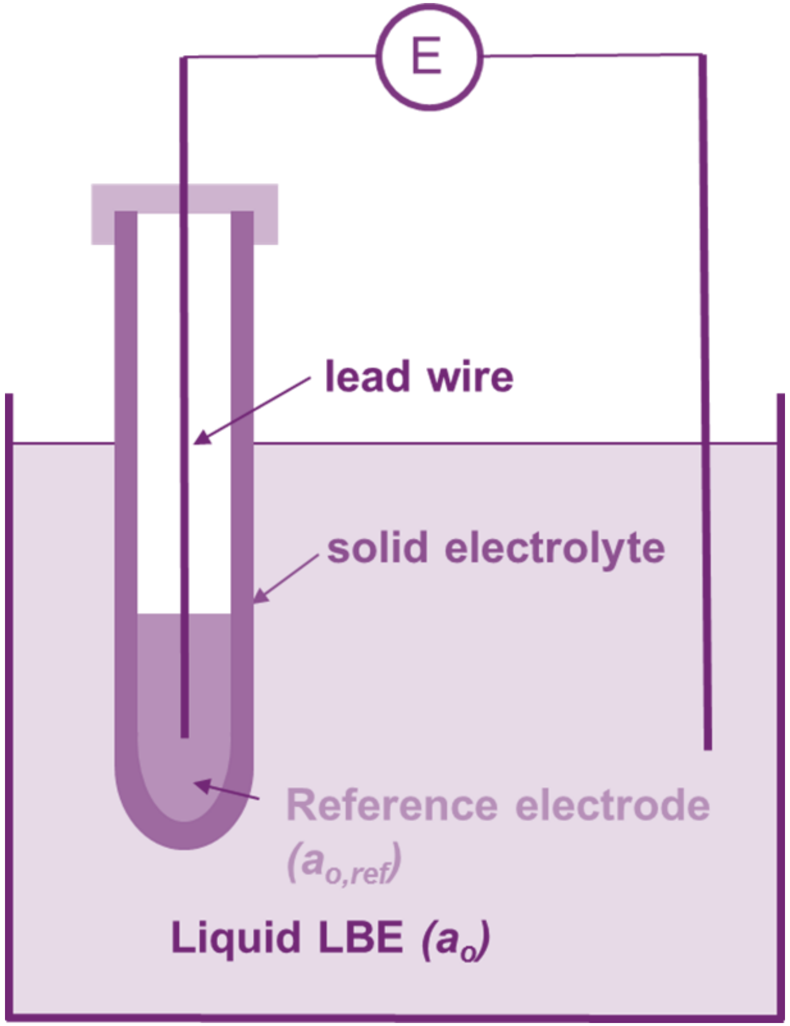
Multi-physics LBE Coolant Chemistry Code

1

Oxygen Sensor

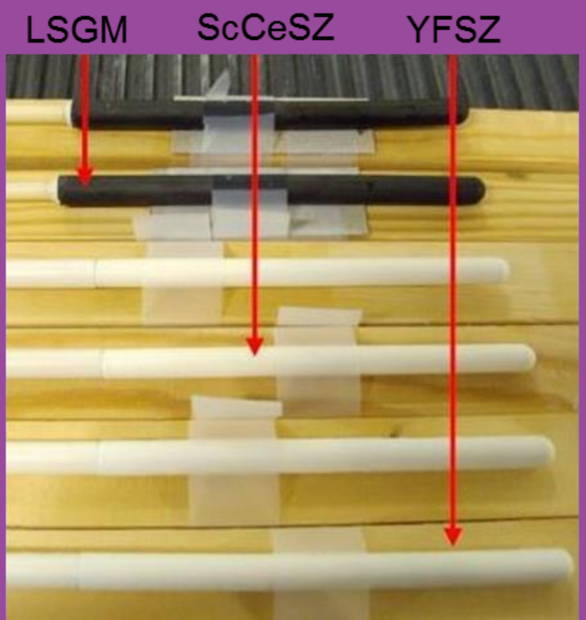

SCK CEN/98311810

$$E = \frac{RT}{2F} \ln\left(\frac{a_{o(ref)}}{a_{o(LBE)}}\right)$$



Reliability

- Solid electrolyte
  - Conductive to O^{2-} ions
  - Not conductive to e-
- Ceramic to metal sealing
- Thermal shock



Temperature range

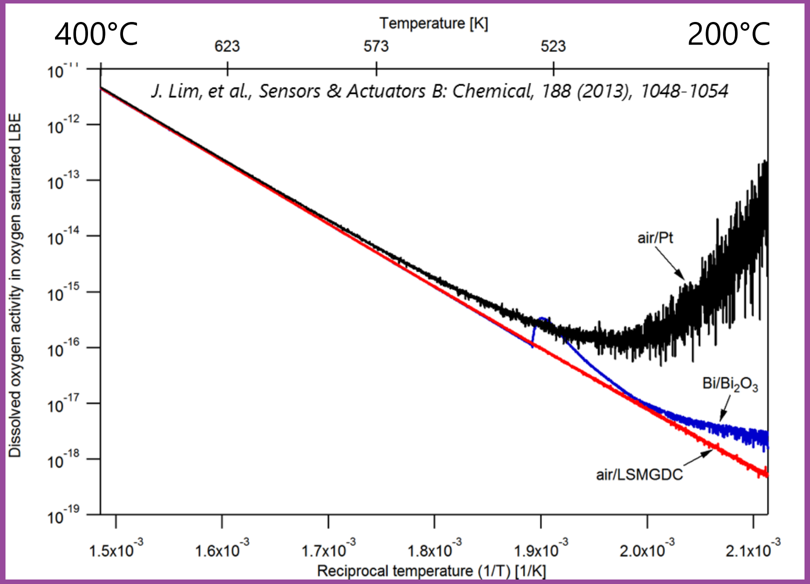
- Differential expansion
- Higher temperatures lead to more stable signals
- Challenging below 350°C
- Target: 200°C

Design and validation

- Improved ceramic/steel sealing and coupling
- Solid electrolyte with superior mechanical properties
  - YPSZ
  - Longer sensors
- Simple but reliable sensor validation method
  - Equilibrium

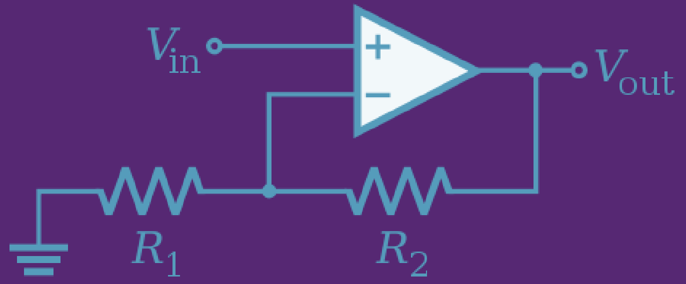
Reference electrode

- New (oxide) electrode
- No coating necessary



Potential measurement

- In-house designed voltage buffer



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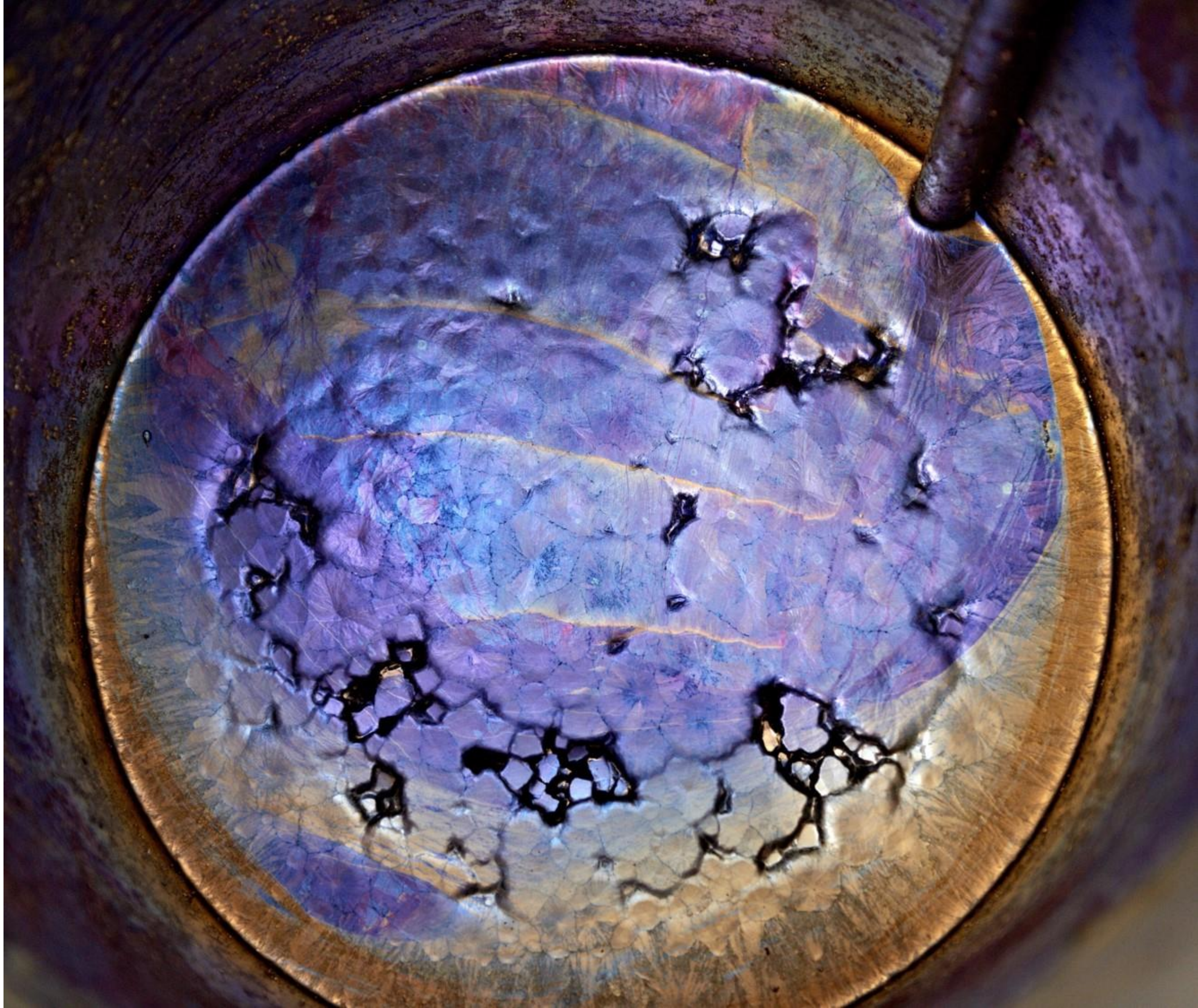
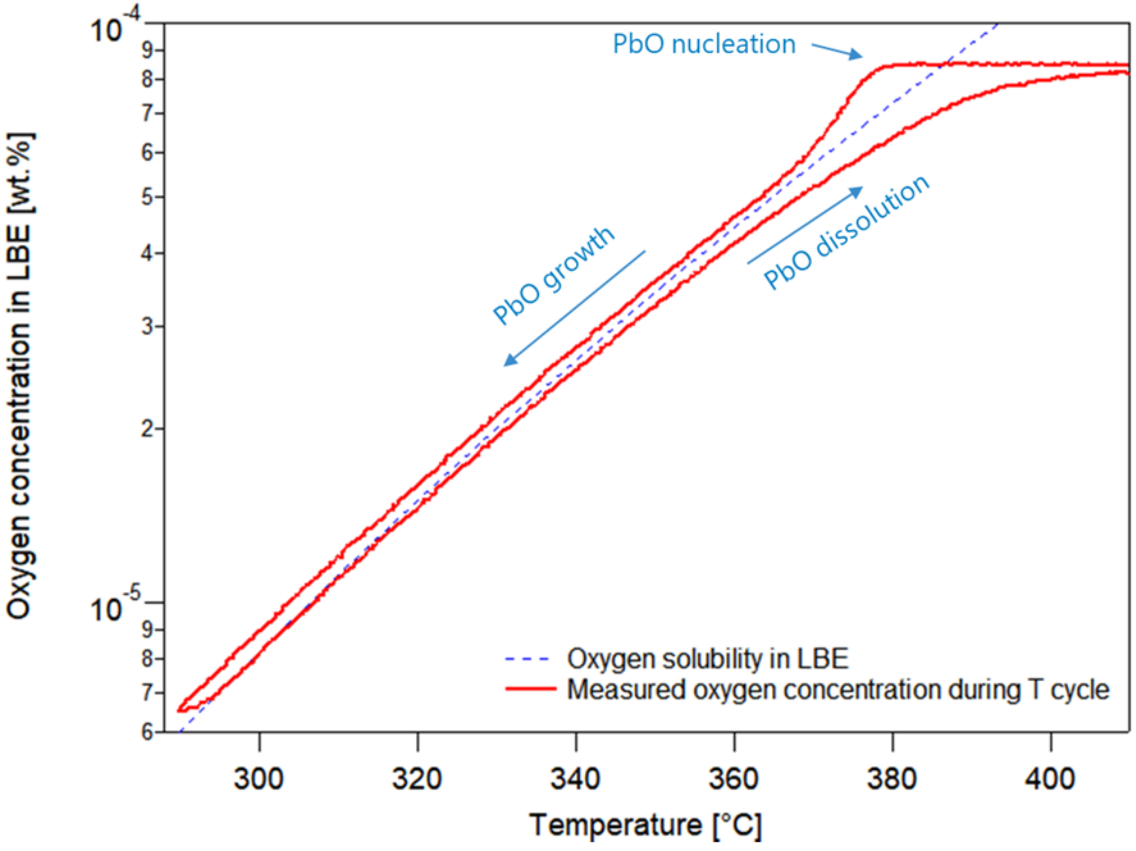
LBE-O Interaction

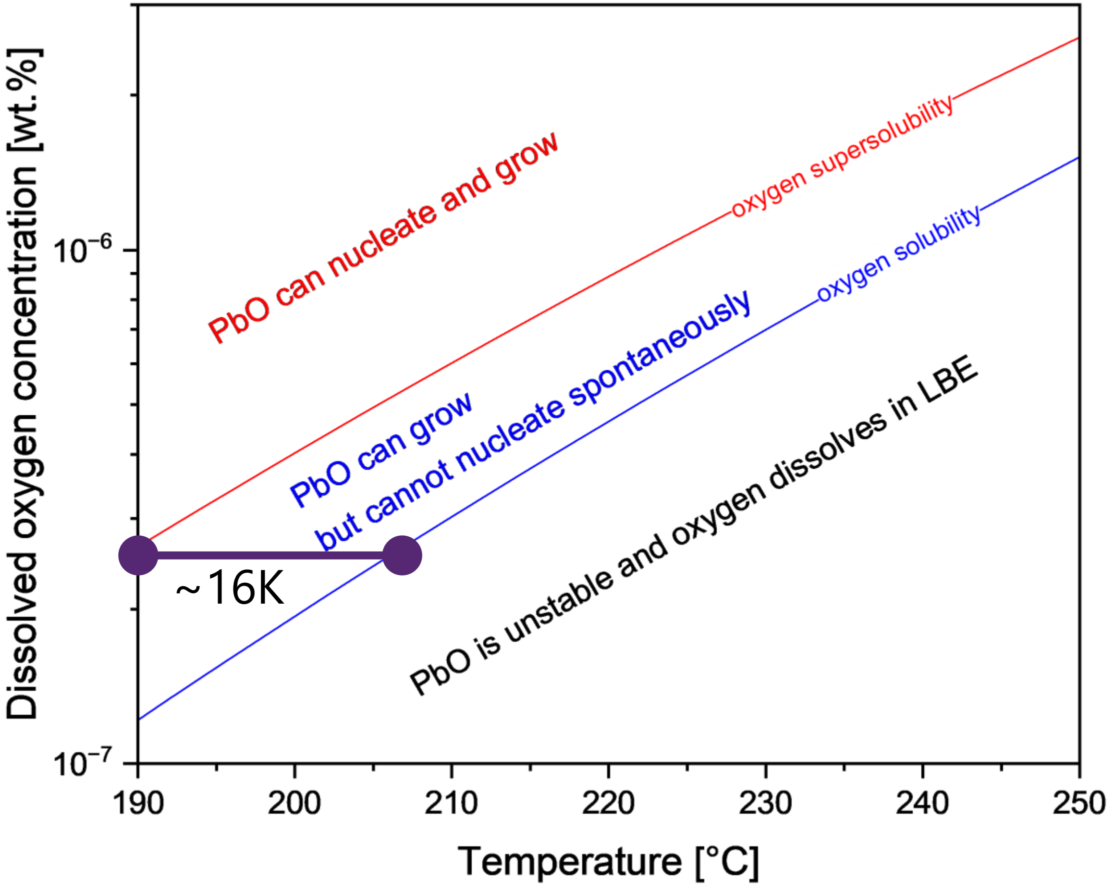


Kitchen example: Sodium Acetate: CH_3COONa (food ingredient: **E262**)

LBE-O : PbO nucleation and growth

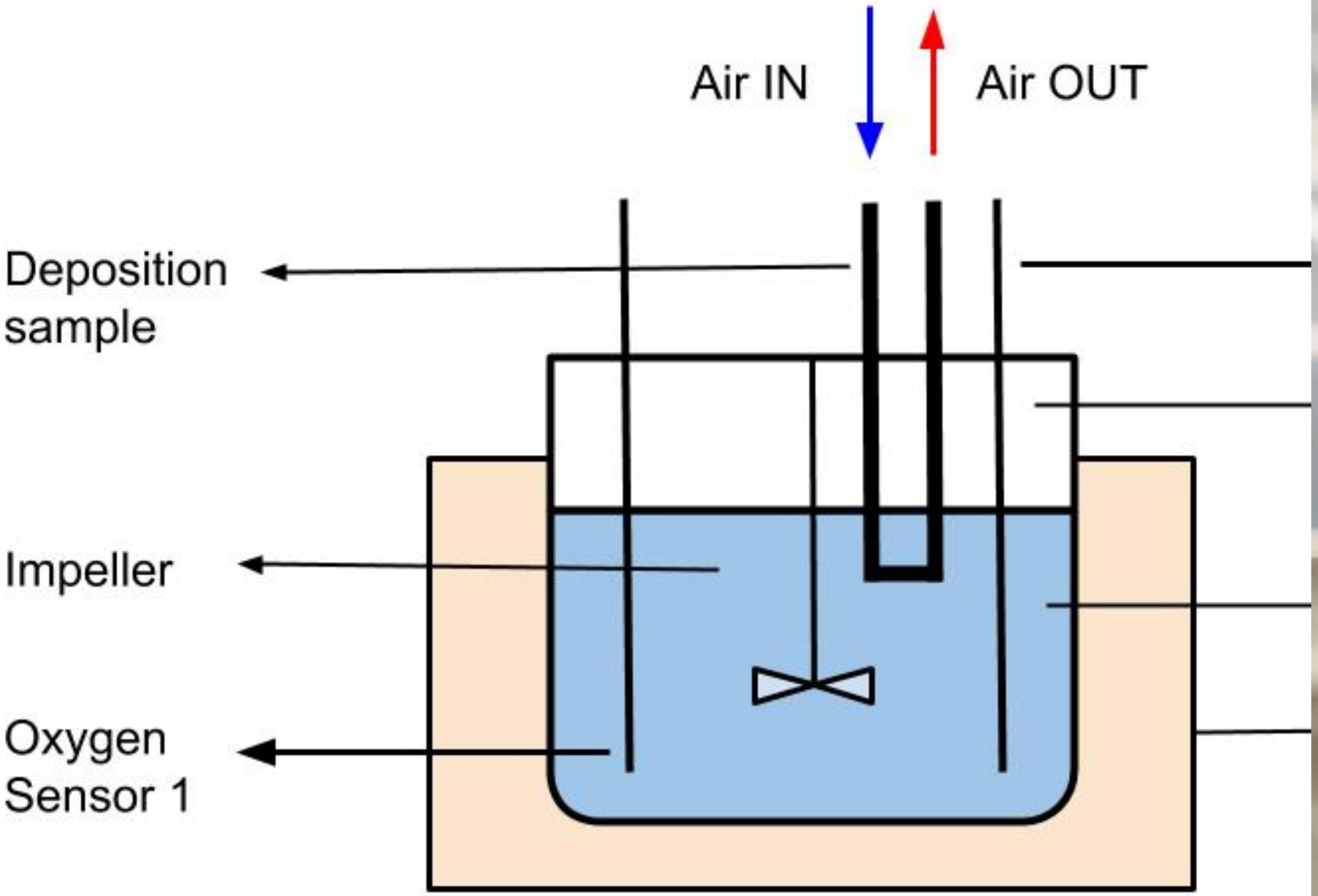


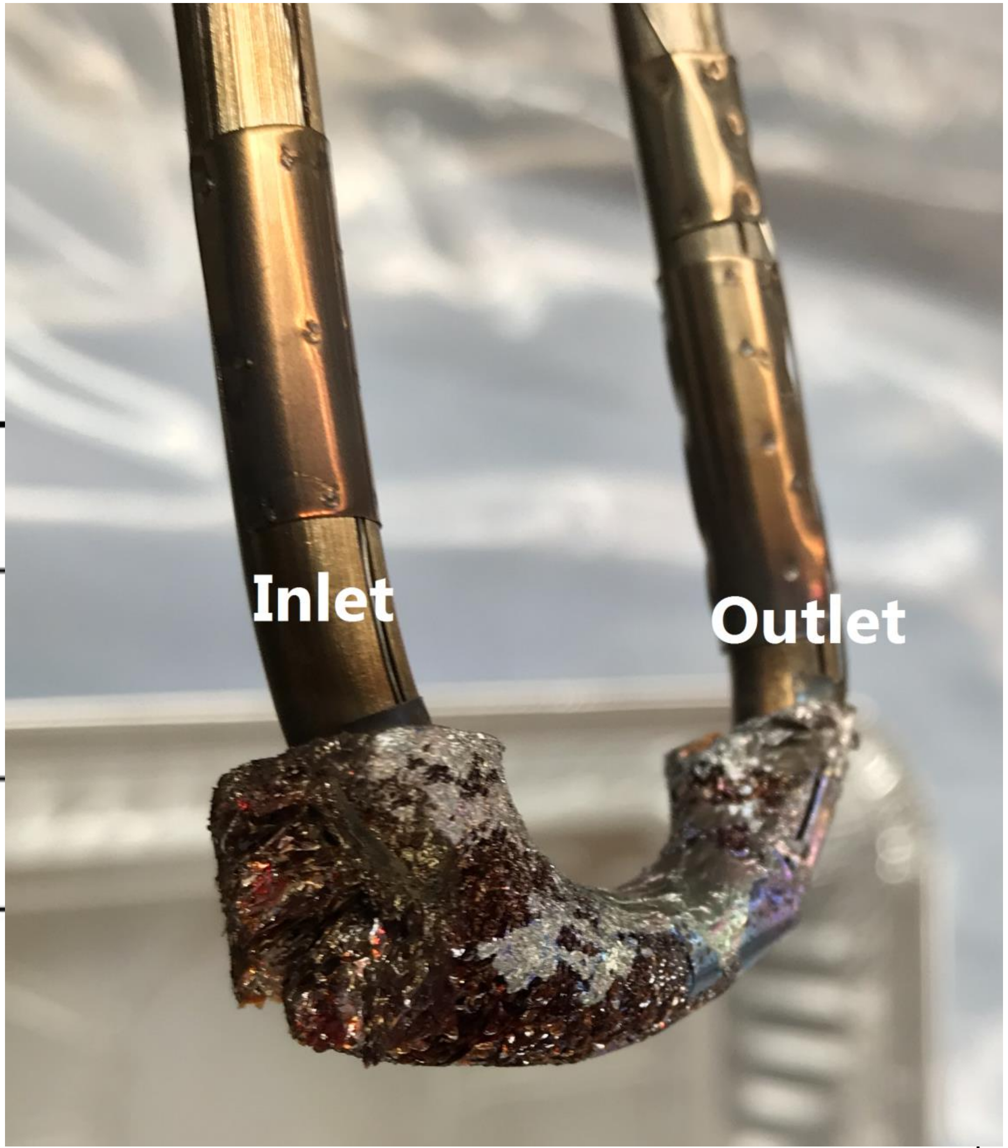
PbO nucleation, growth and dissolution in LBE

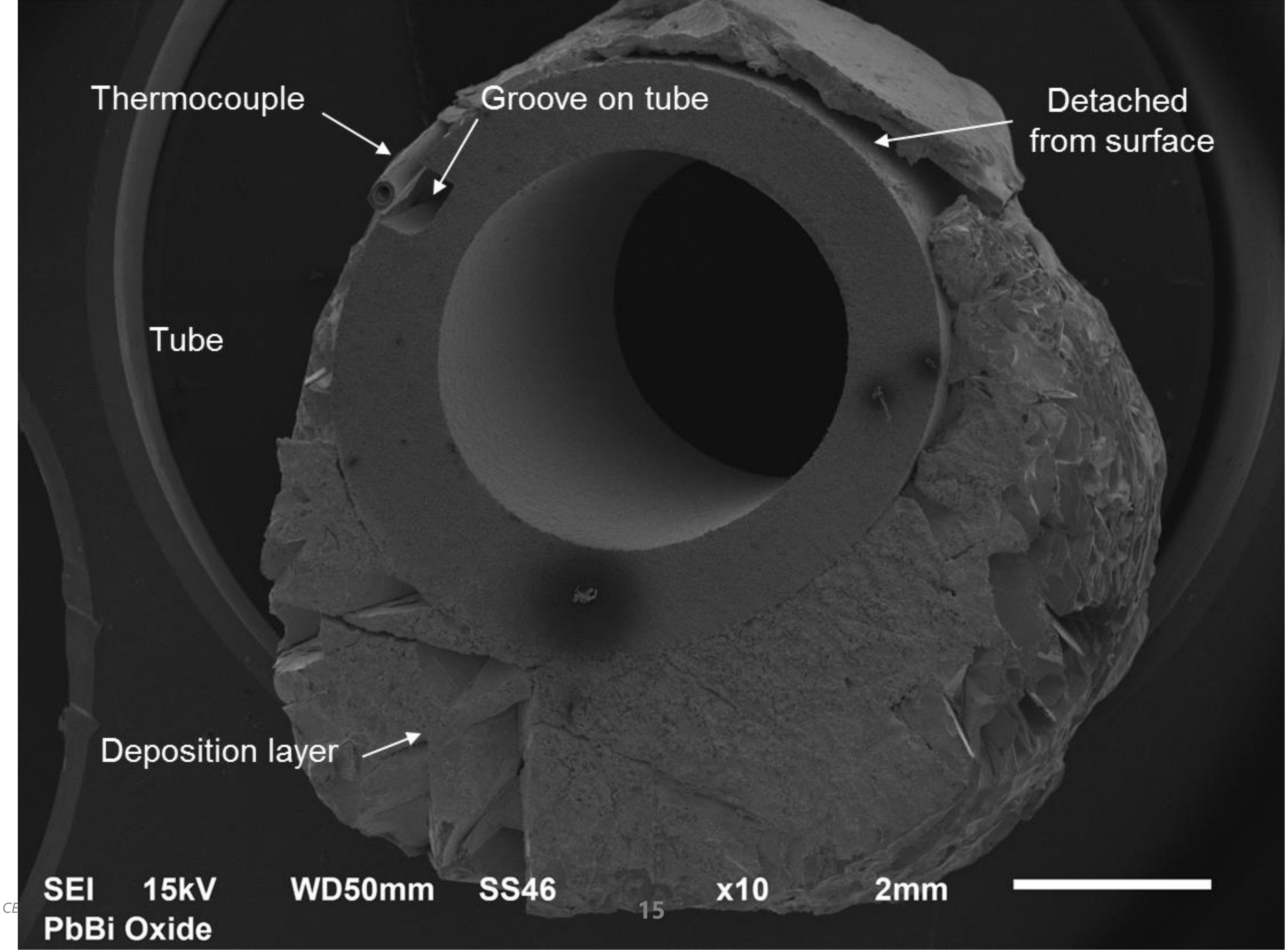


- J. Lim, et al., JOM, 73 (2021) 4023-4029
- K Gladinez, et al., Physical Chemistry Chemical Physics 19 (2017), 27593-27602

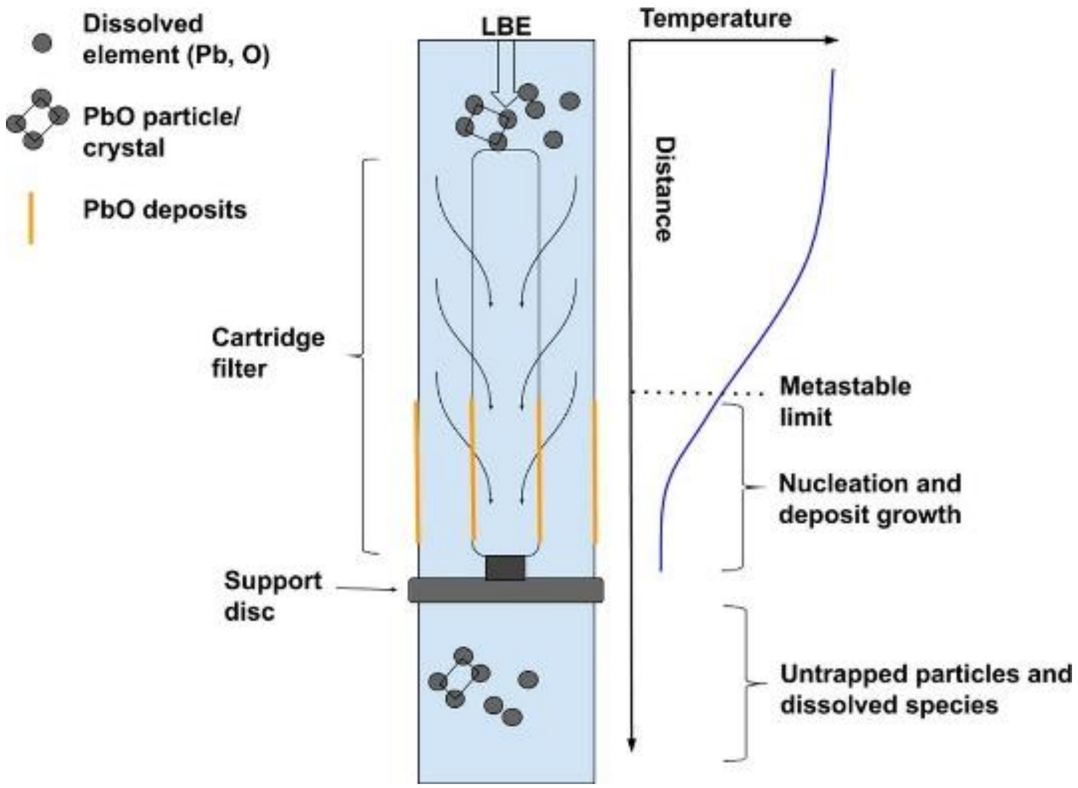
Cold finger setup OSCAR

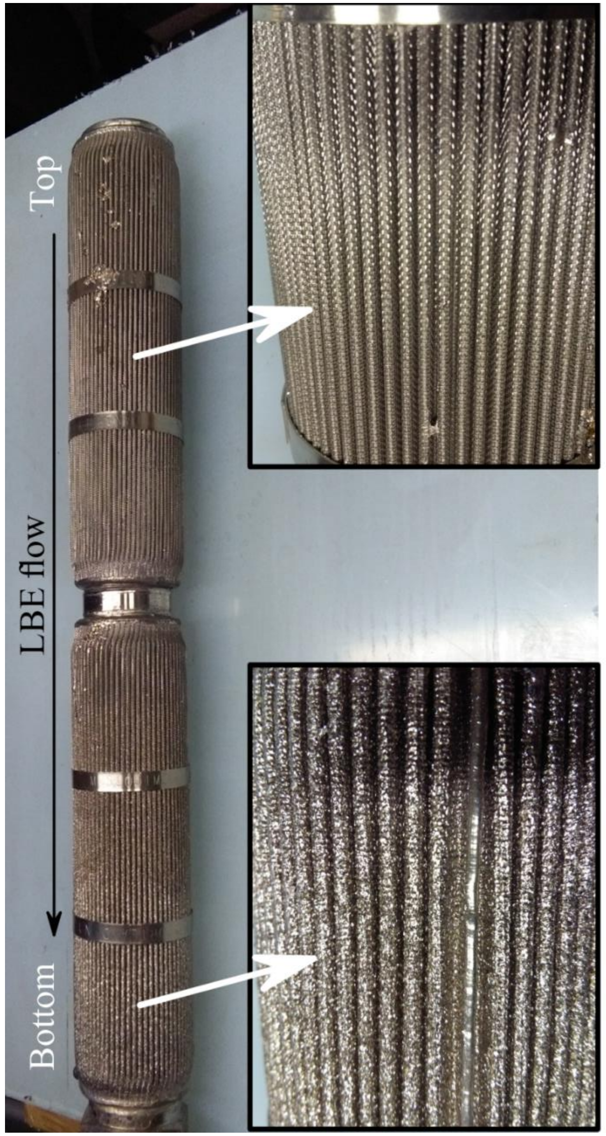


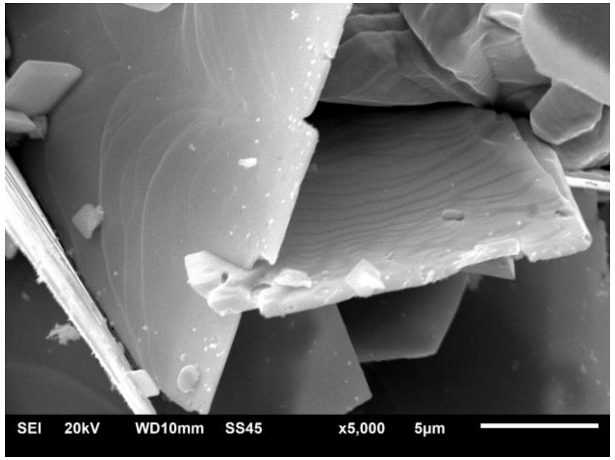


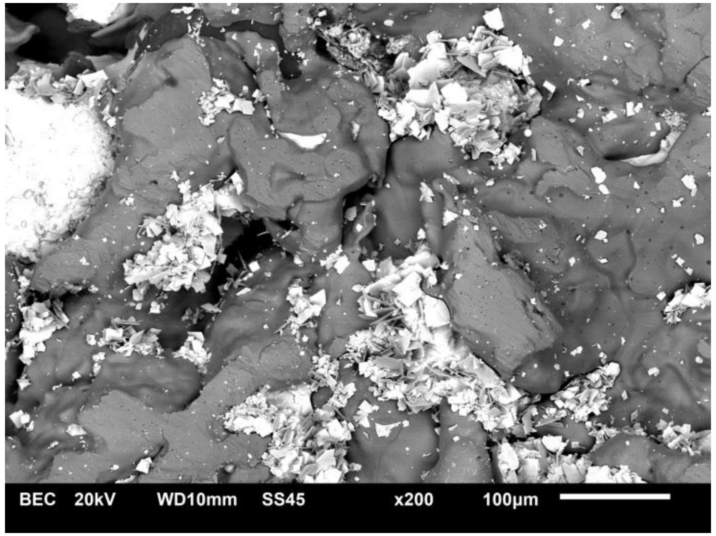

Thermocouple
Groove on tube
Detached from surface
Tube
Deposition layer
SEI 15kV WD50mm SS46 x10 2mm
PbBi Oxide

LBE-O interaction : Cold Trap









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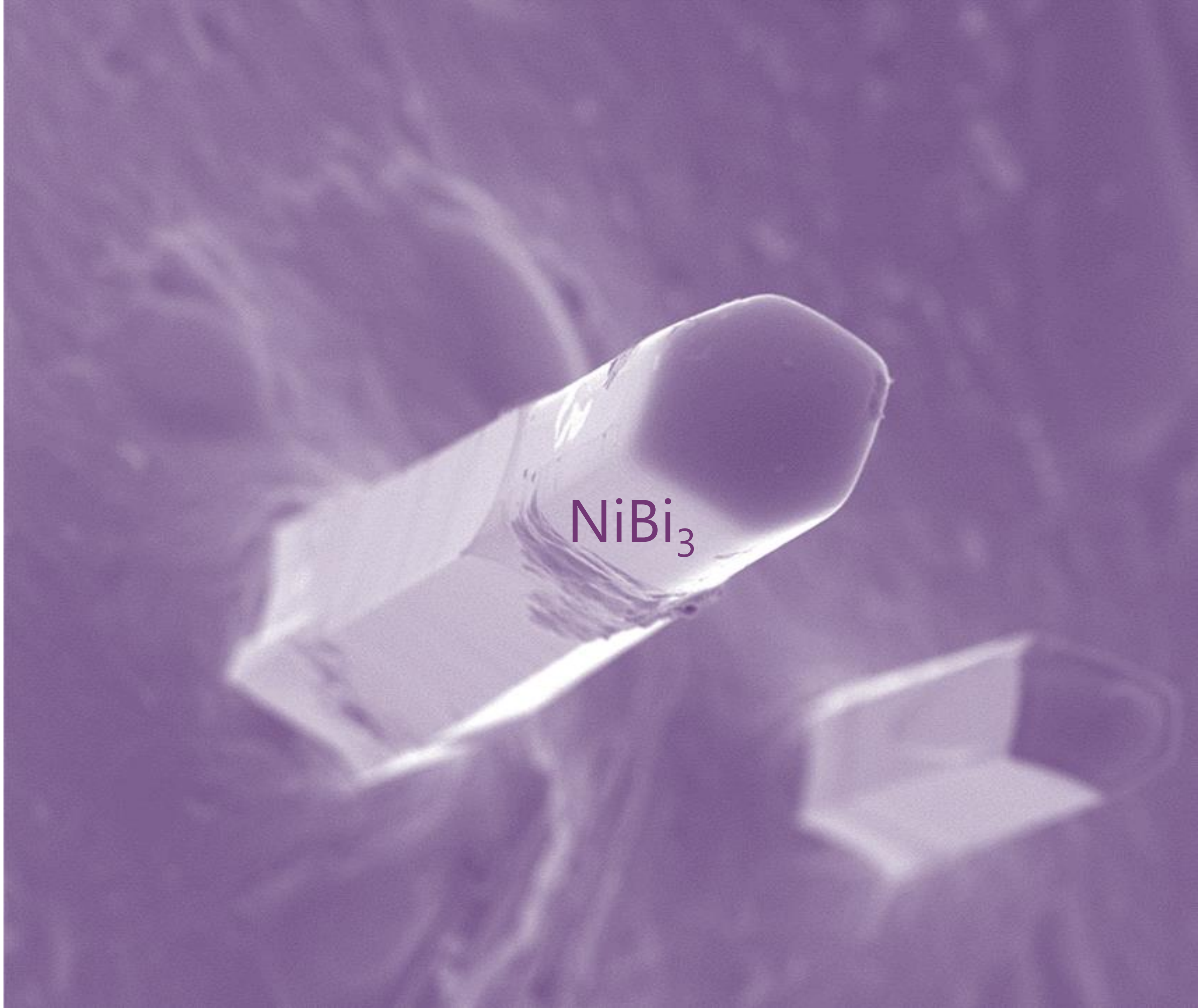
Corrosion Products



Corrosion products : Fe, Cr, Ni, Co

Fe

$Fe_{(lbe)}$
Fe_3O_4
Fe_2O_3

Ni

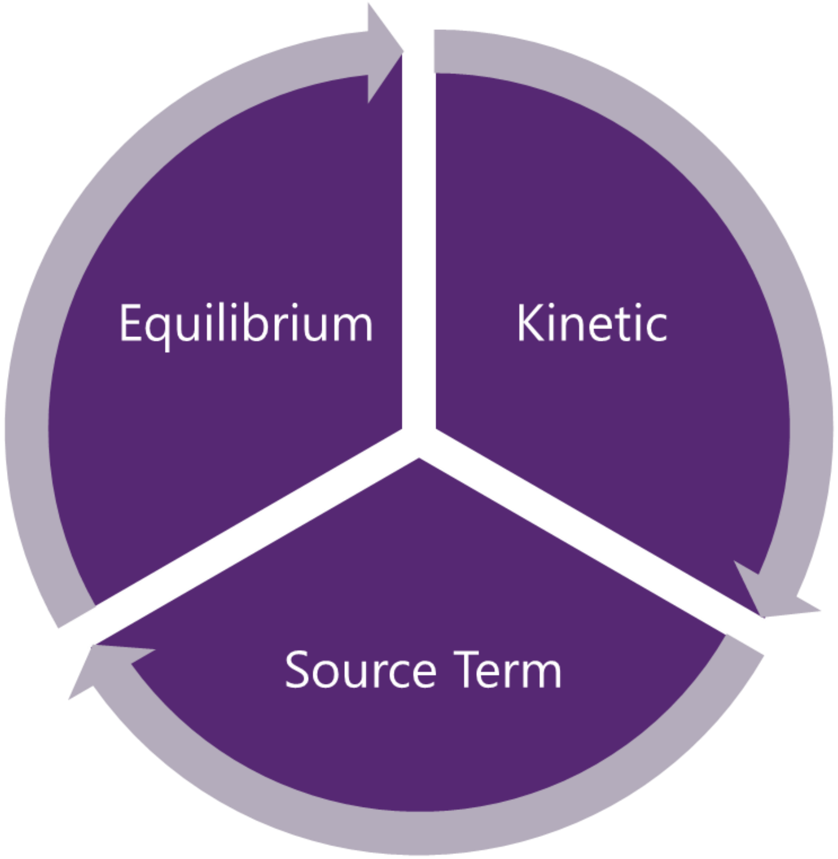
$Ni_{(lbe)}$
NiO
$NiBi_3$

Cr

Cr_2O_3

Co

$Co_{(lbe)}$
CoO



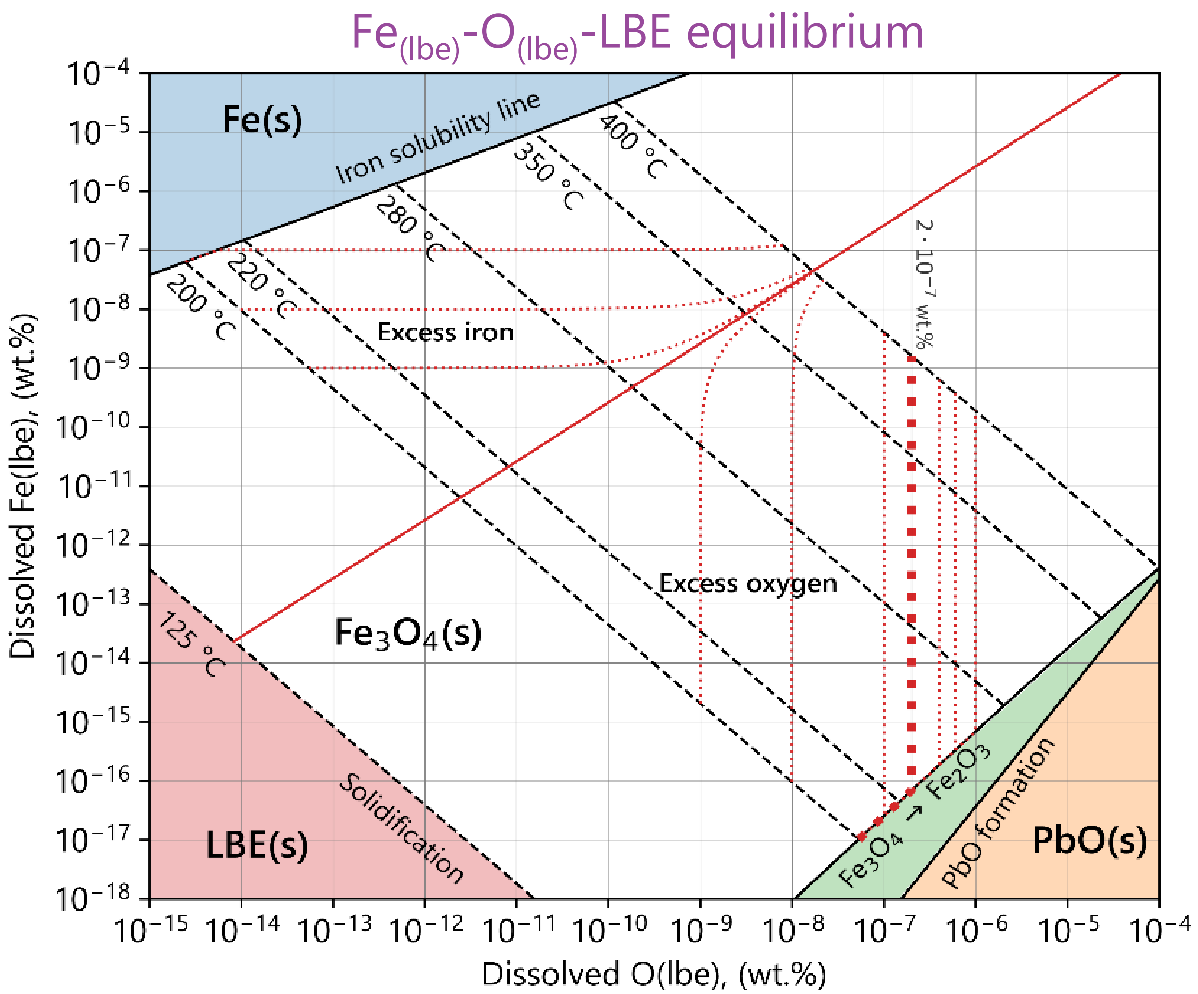
Corrosion products : Source term estimation

Assumption & Limitation :

- *Uniform corrosion*
- *Release rate from corroded area : Fe (10%), Cr (50%), Ni (100%), Co (100%)*
- *Corrosion depth estimated based on data from stagnant, low oxygen, isothermal LBE*

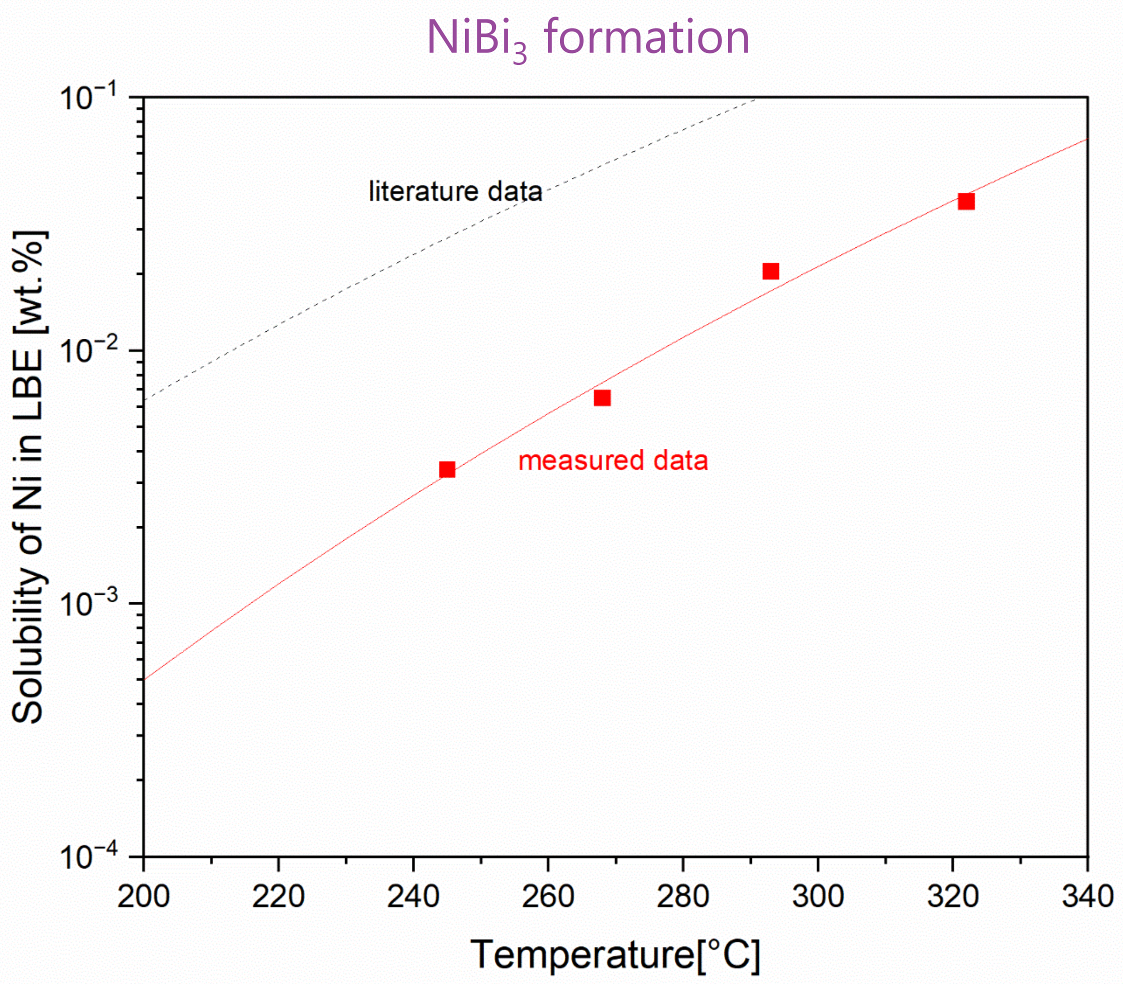
	Fe	Cr	Ni	Co	**Total**
Release rate [g/h]	0.06	0.07	0.10	0.0003	**0.23**
40 years [kg]	16	18	26	0.08	**60**
40 years [ppm]	3.2	3.6	5.2	0.016	**12**
Initial impurities [ppm] (as received, comercial non-nuclear grade)	<2	trace	<4	<1	**Ag: 21** **Cu: 7** **Hg: <2**

Corrosion products: Fe



- Fe is the corrosion product that has significant impact on LBE coolant chemistry.
- Oxygen control range of MYRRHA falls within excess oxygen region.
- Magnetite/Hematite transition could potentially impact on the oxygen chemistry in MYRRHA

Corrosion products: Ni



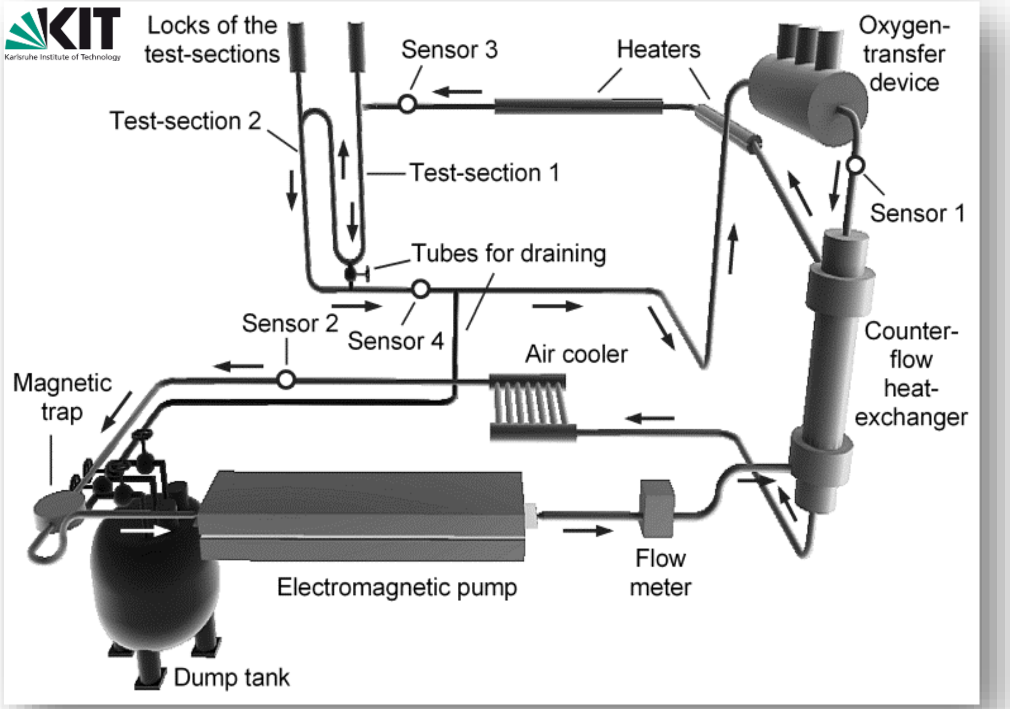
- Ni is expected to be the largest amount of corrosion product.
- Approximately 5 ppm of nickel will accumulate in the coolant, in addition to the initial Ni impurity.
- $NiBi_3$ could start to precipitate after 20 years operation.
- Cold trapping of Ni in the form of $NiBi_3$ is necessary.

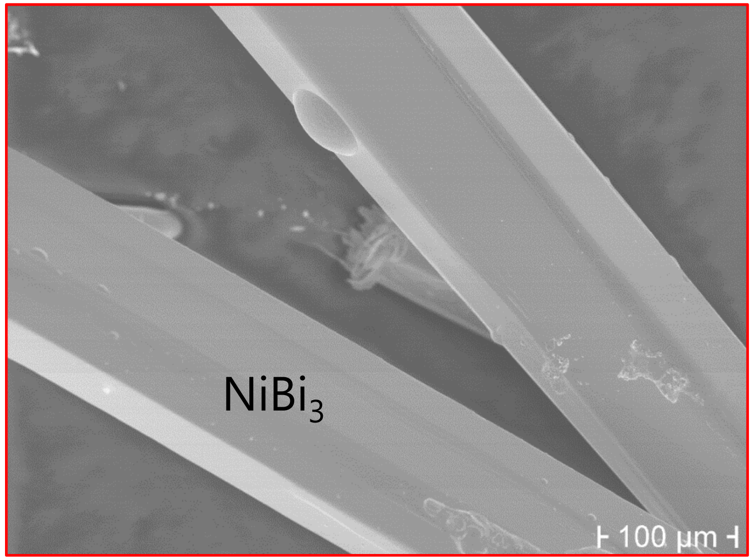
- G. Gregori, et al., J of nuclear materials, 589 (2024) 154866

Learning from experience (CORRIDA loop)

- Cold leg = 400°C, hot leg = 550°C → mass transfer of corrosion products

LBE flow blocked after
113000 hours (~13 years)



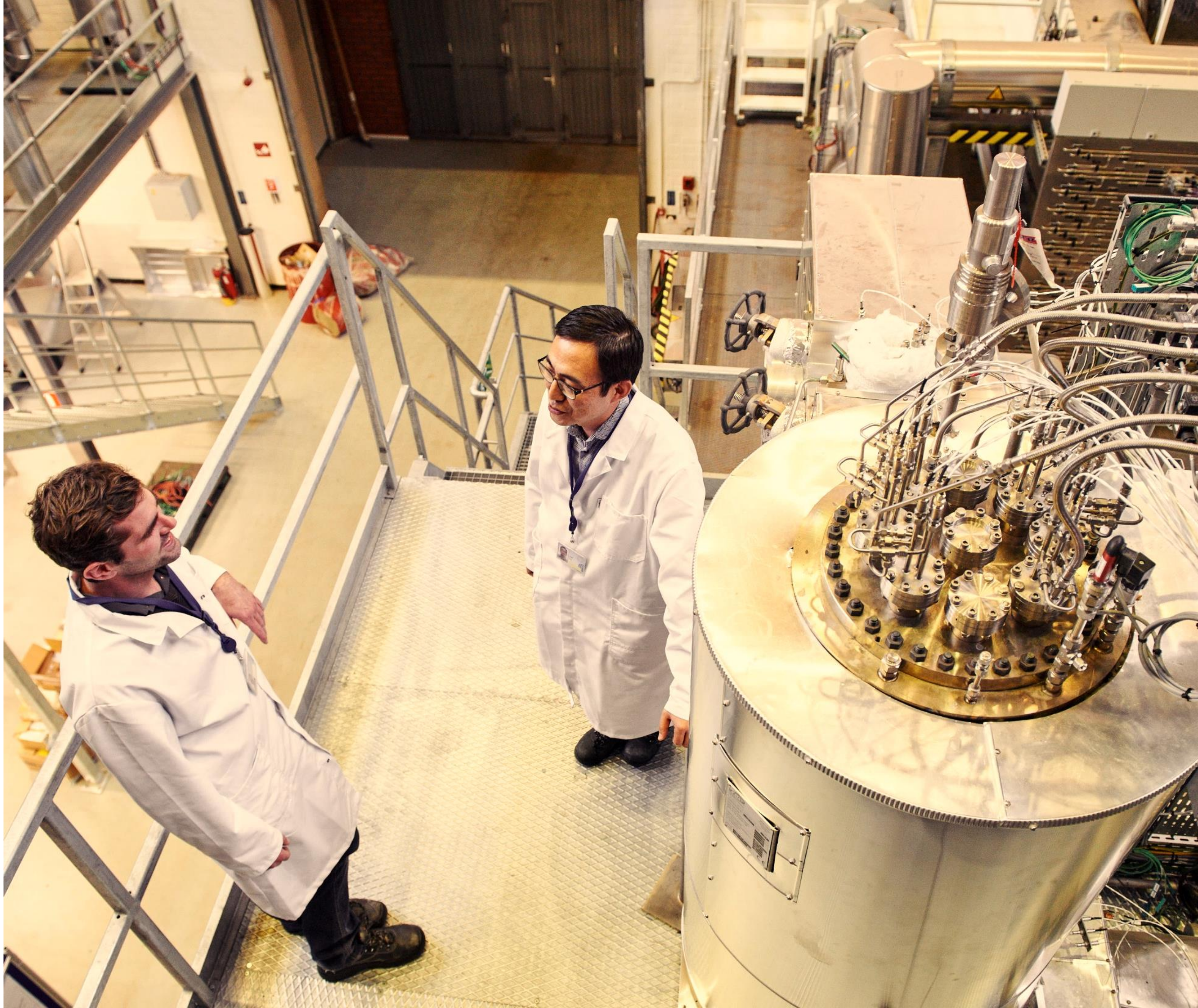


Learning from experience (CRAFT)

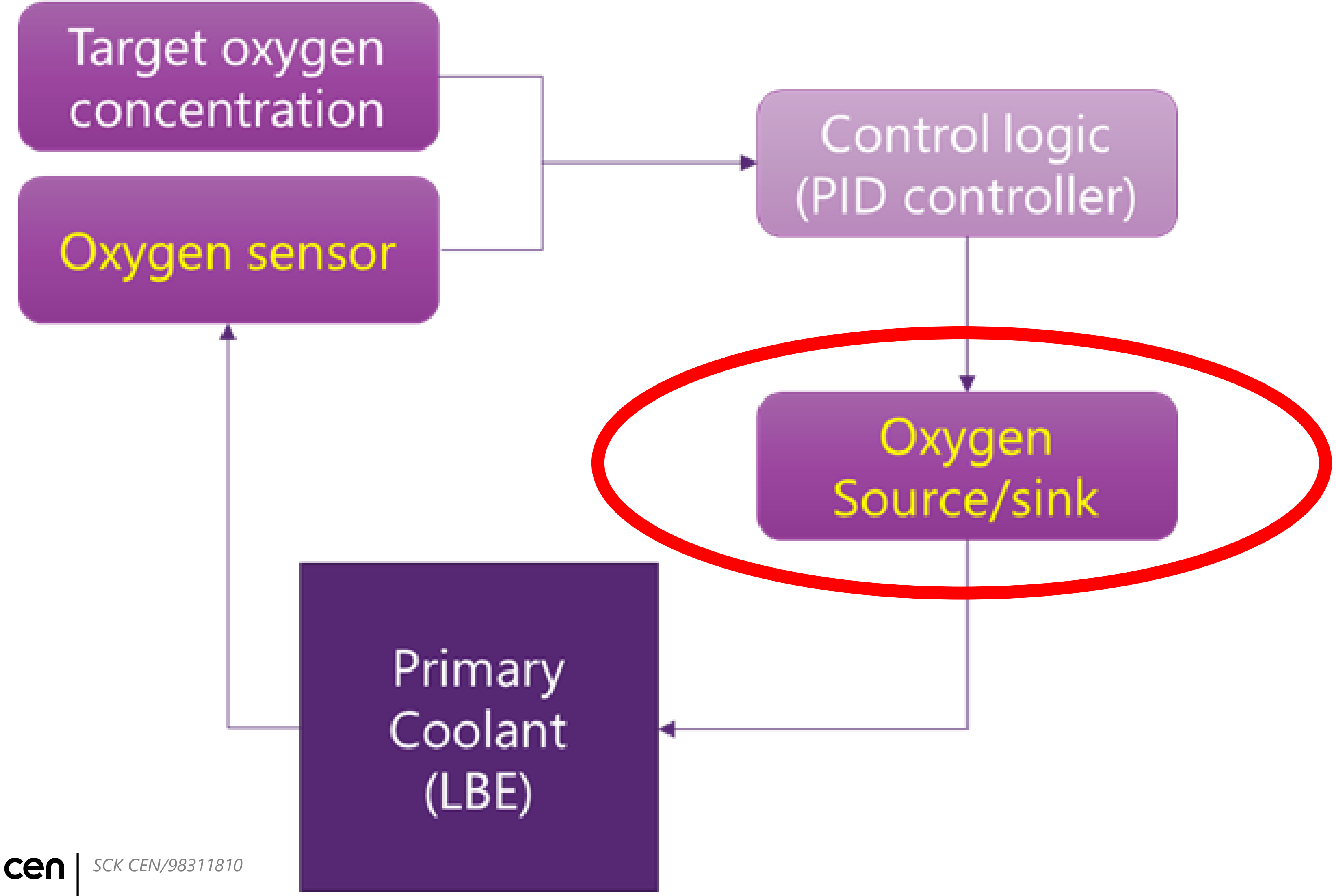
- ✓ Pump motor vibrations
- ✓ Pump power oscillations at high power levels
- ✓ Pump power quick raise
- ✓ Freezing of pressure gage tubes
- ✓ Samples disconnection during operation
- ✓ Tracer heaters failure of the draining line
- ✓ Leakage in the economizer
- ✓ Crack of welded joint in P22 steel
- ✓ Flow meter failure

4

Oxygen Control



Oxygen Control Method

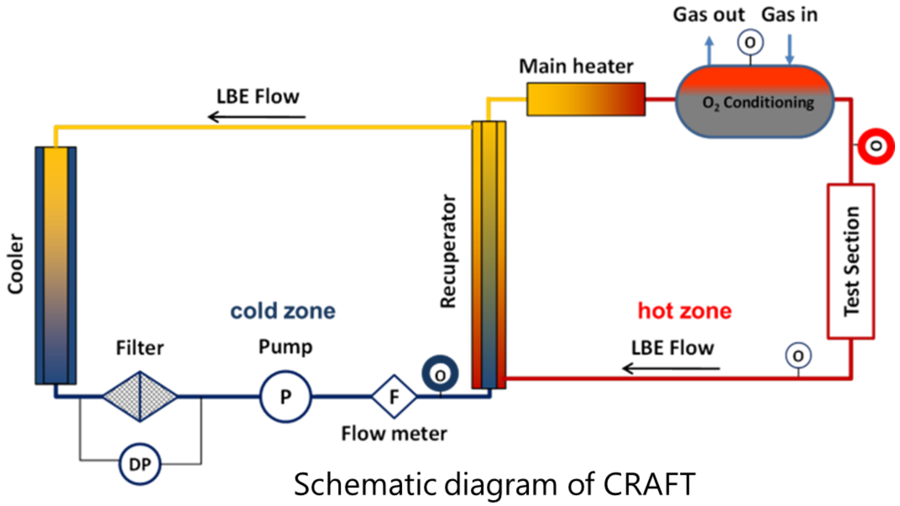


Oxygen Control System Validation

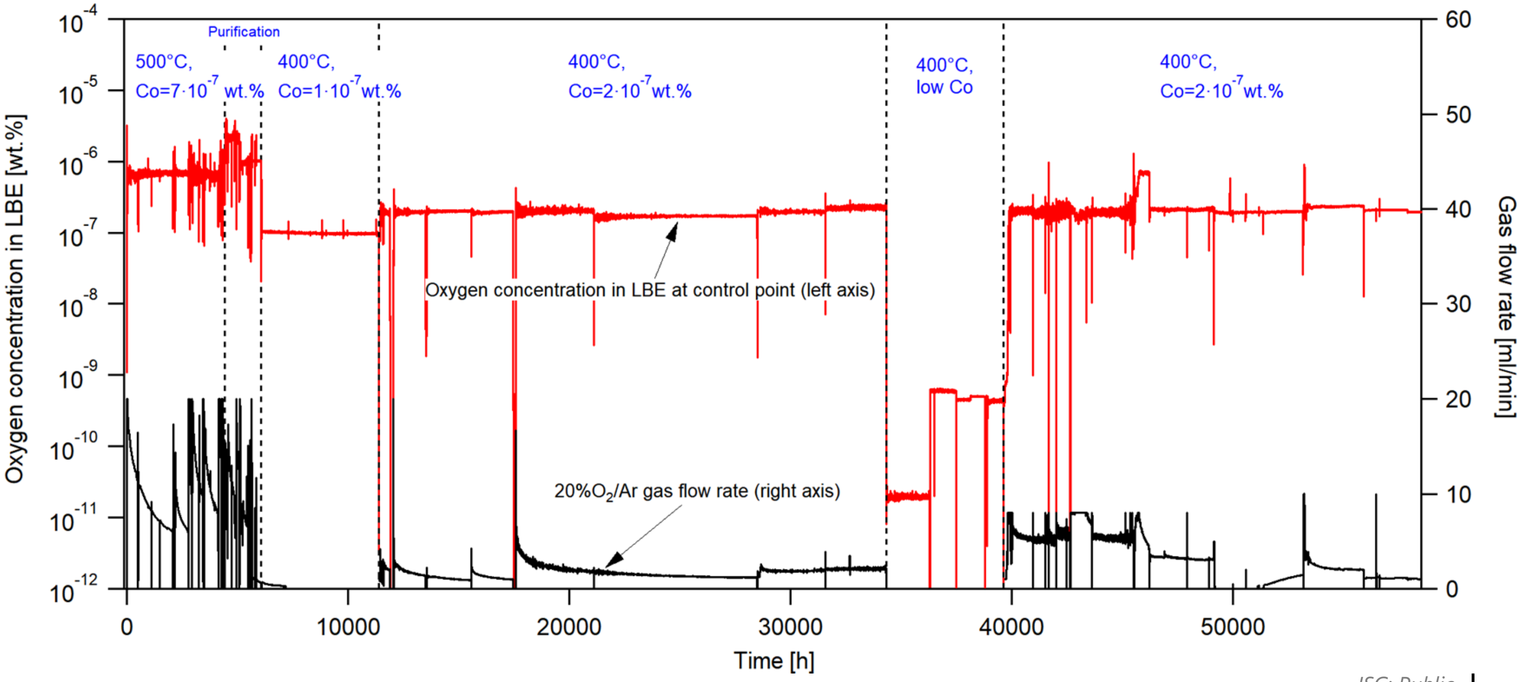
	Technology	Chemical Reactions	SCK CEN test facility
	Gas control	$O(lbe) + H_2(g) \rightarrow H_2O(g)$ $O_2(g) \rightarrow 2O\ (lbe)$	Helios 3 pool (bubbling) CRAFT corrosion loop (cover gas)
	PbO mass exchanger (PbO MX)	$PbO(s) \rightarrow Pb(lbe) + O(lbe)$	MEXICO chemistry loop
	Electrochemical oxygen pump (EOP)	$2O^{2-}(lbe) \rightarrow O_2\ (g) + 4e^-$ $O_2\ (g) + 4e^- \rightarrow 2O^{2-}(lbe)$	MEXICO chemistry loop
	Cold trap	$Pb(lbe) + O(lbe) \rightarrow PbO(s)$	MEXICO chemistry loop
	Oxygen getter	$xM(s) + yO(lbe) \rightarrow MxOy(s)$	MEXICO chemistry loop

Cover gas control in CRAFT

- Active control of 20% O_2/Ar flow rate under fixed 5%H_2/Ar and Ar flow

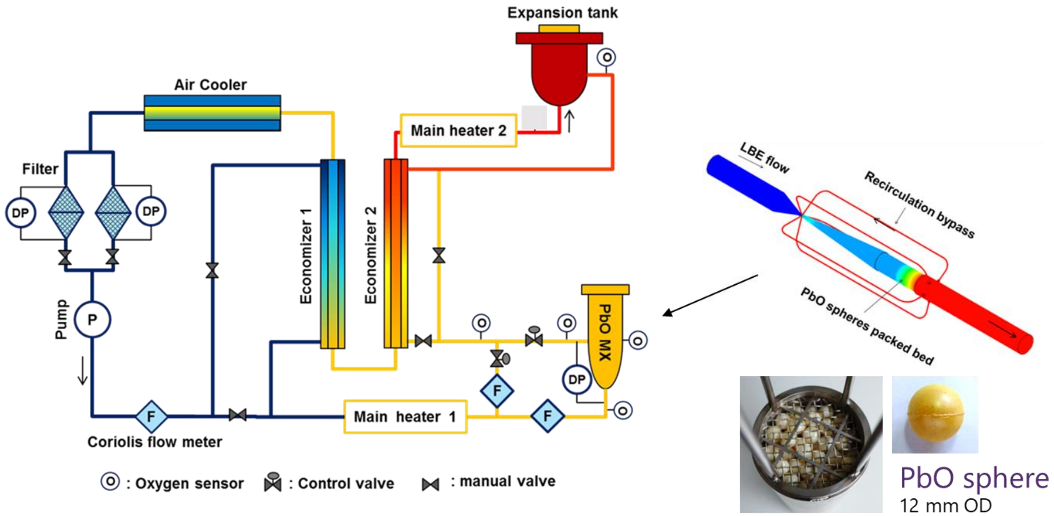


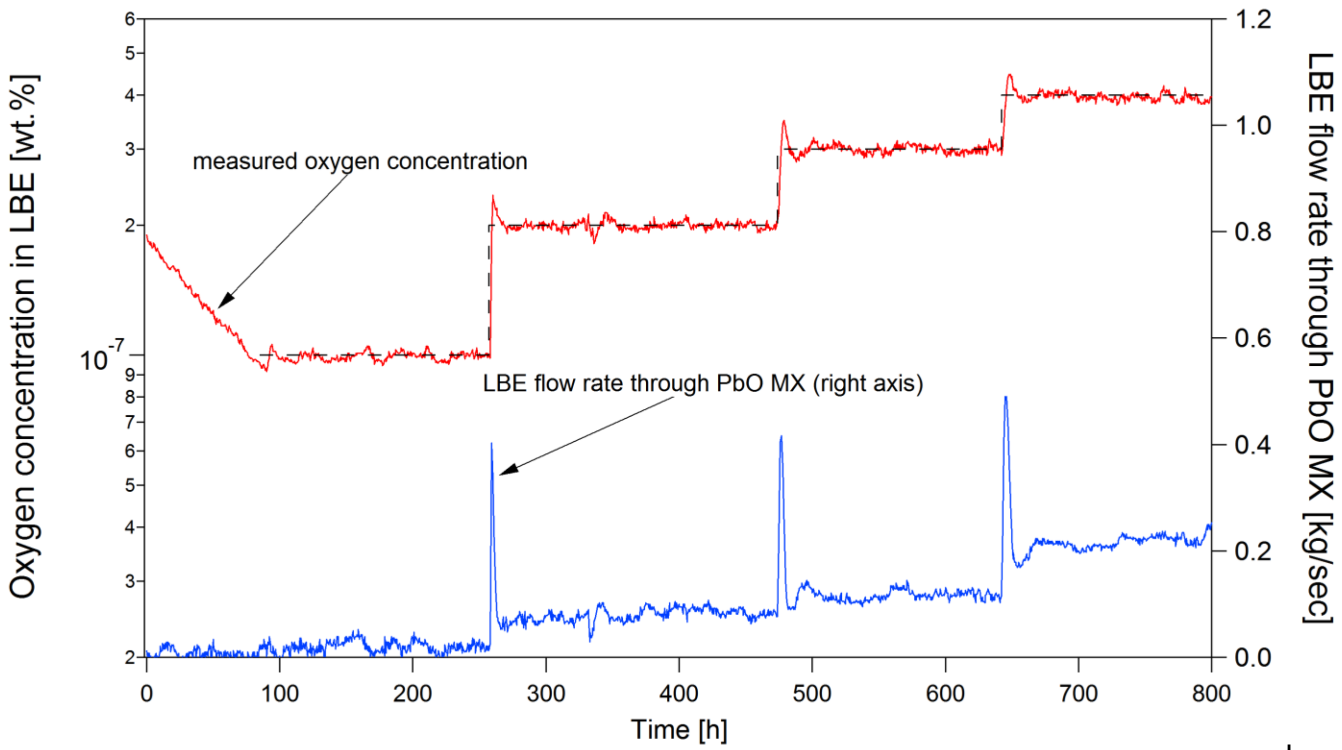
Schematic diagram of CRAFT



PbO MX: MEXICO Loop

- Automated control of local flow rate over PbO MX by regulating control valves

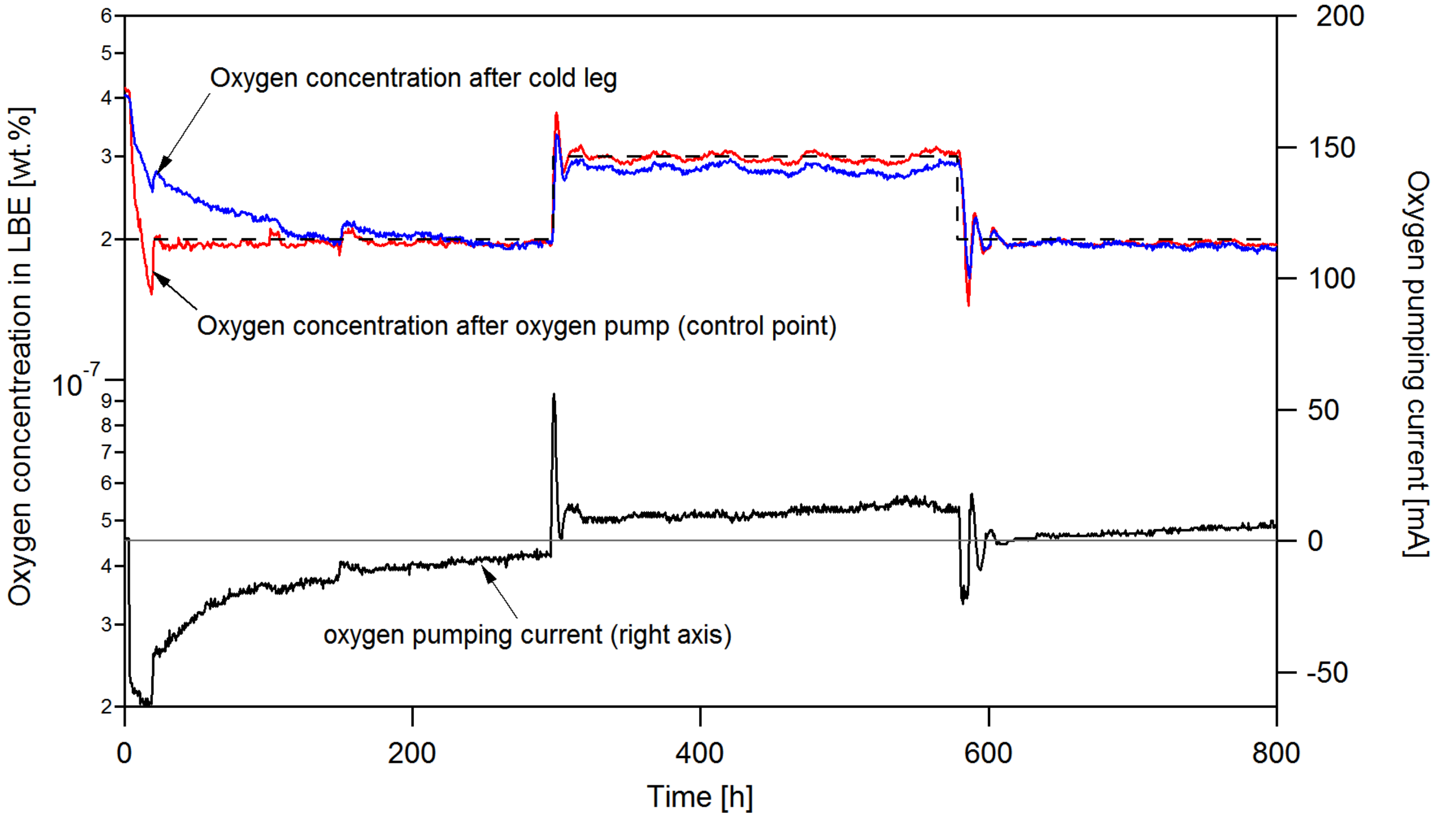




Electrochemical Oxygen Pump in MEXICO

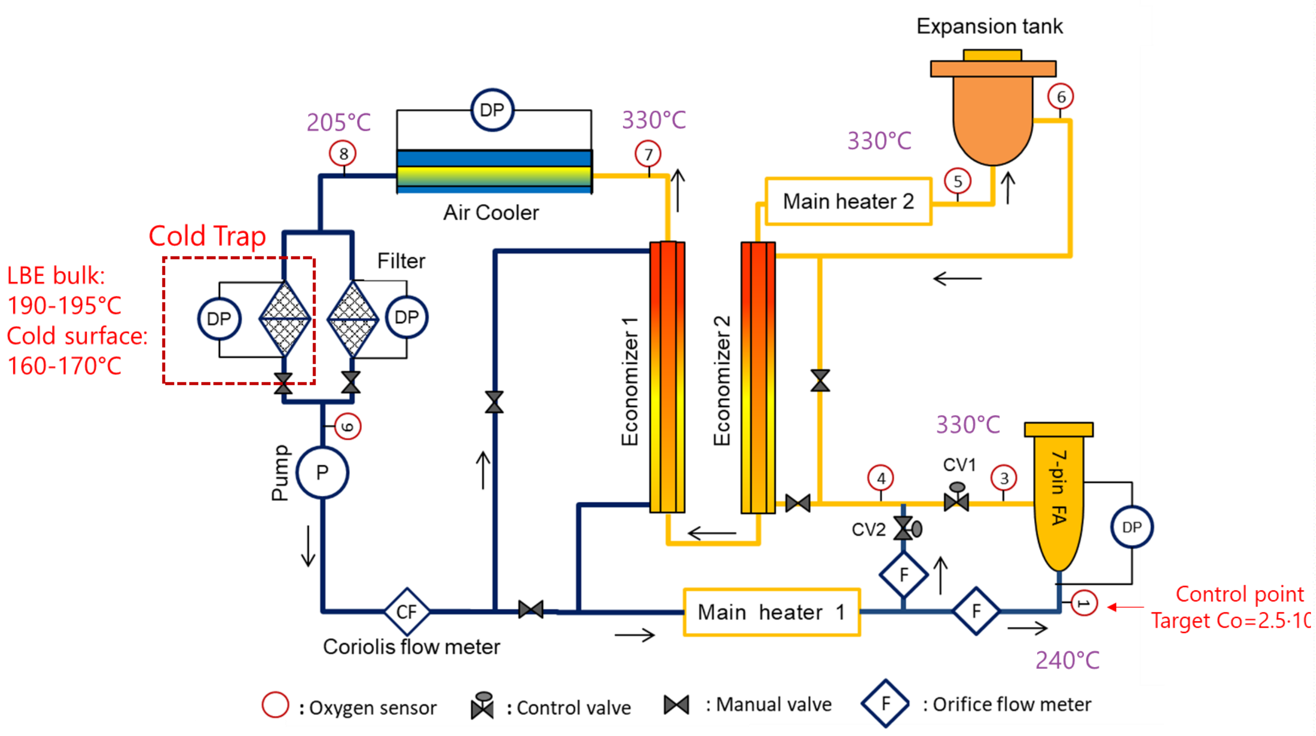
- Automated control of electrical current over EOP

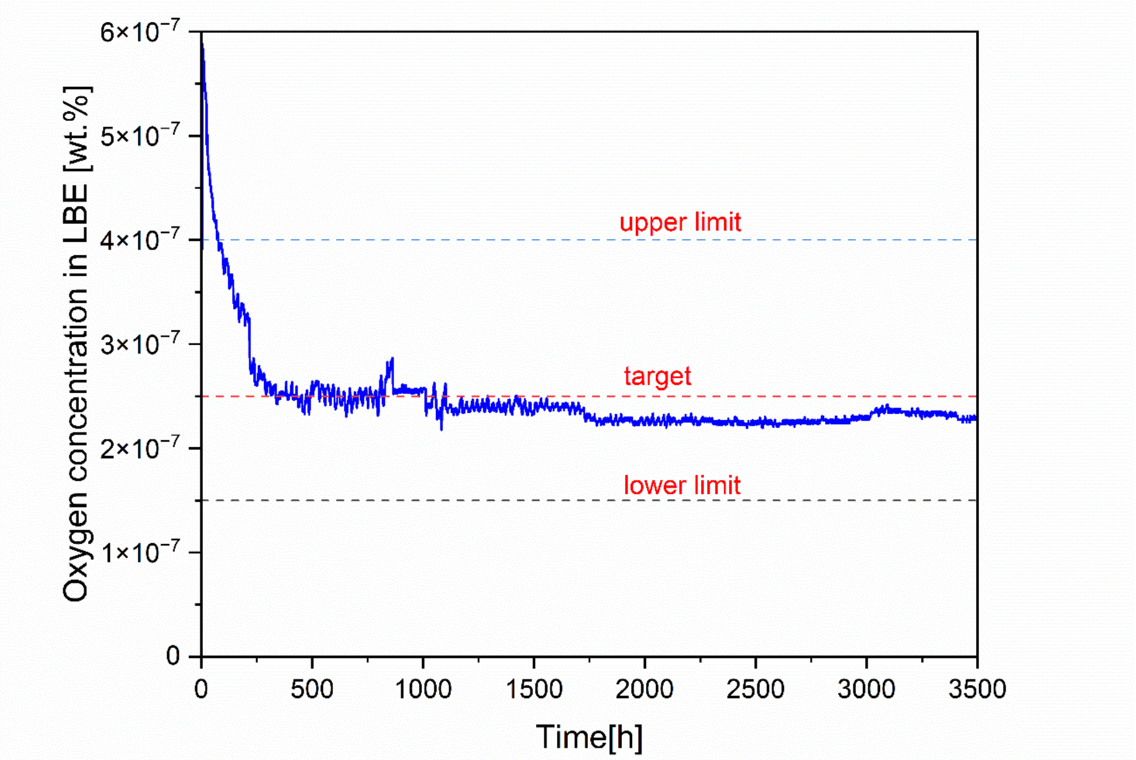
Oxygen pumps installed in MEXICO
(5 modules, 15 pumps in operation)



Cold Trap in MEXICO

- Control temperature in Cold Trap



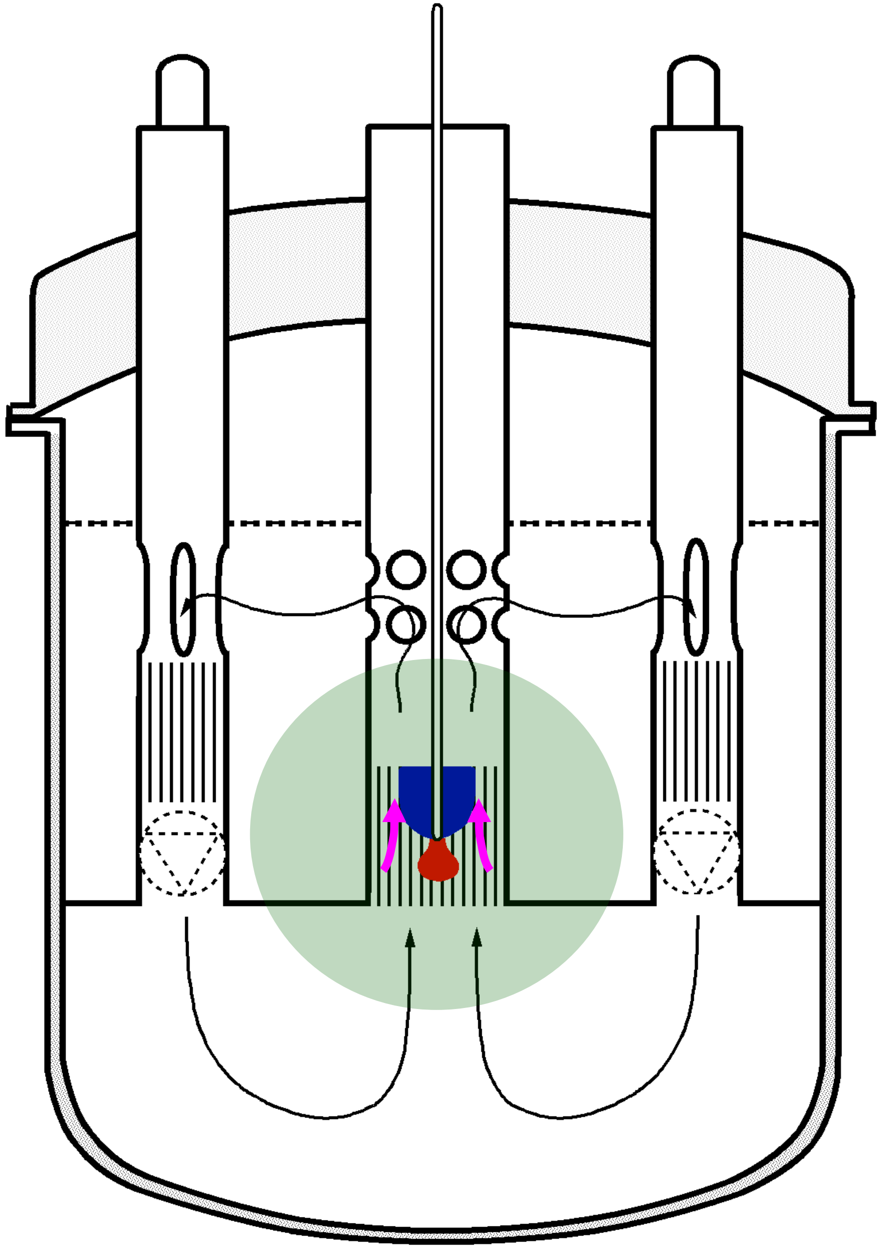


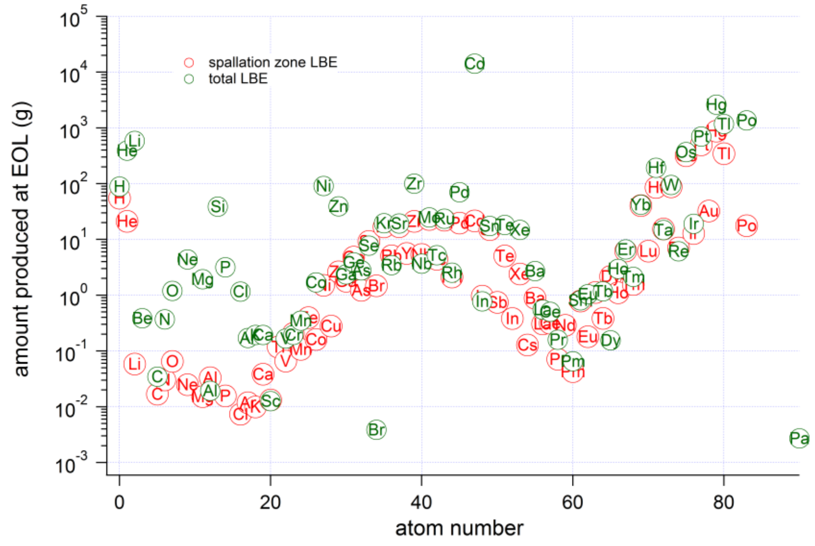
Cold Trap & PbO MX have been chosen for MYRRHA

Methods	Operating Temperature	Characteristics
Gas phase oxygen addition	>250°C	• Gases are readily available (+) • Simple and easy to control (+) • Possible influence on radionuclides evaporation by H_2 or O_2 gases (-)
Gas phase oxygen removal	>400°C	
PbO MX (oxygen addition)	**>250°C**	• Higher surface to volume ratio (+) • Risk of poisoning PbO (-) • Replacement of PbO bed is necessary (-)
Cold trap (oxygen removal)	**<220°C**	• Efficient oxygen removal at low temperatures (+) • Complex system (-) • Replacement of filters is necessary (-)
Electrochemical oxygen pump (addition/removal)	>400°C	• Simple and easy to control (+) • Difficult to increase interface area (-) • Prone to mechanical failure (-)
Oxygen getter (oxygen removal)	TBD	• Potential to remove oxygen at lower temperature (+) • Risk to contaminate coolant (-)

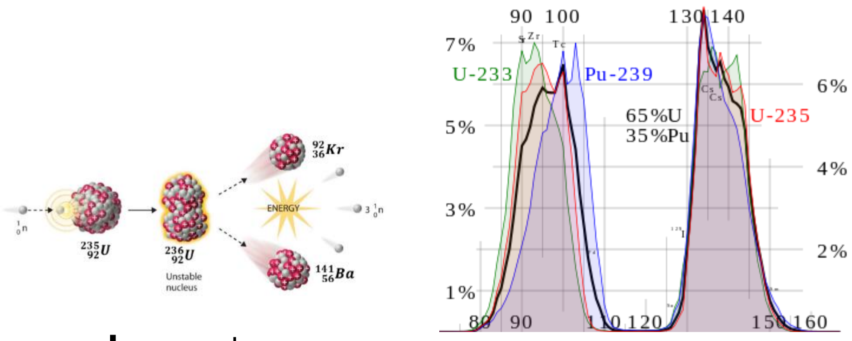
5

Radioactive impurity release





Radioactive impurity sources



SCK CEN/98311810

1 Spallation products

elements generated after protons hit the LBE spallation target (Hg)

2 Activation products

arising from neutron capture reactions with the coolant (Po)

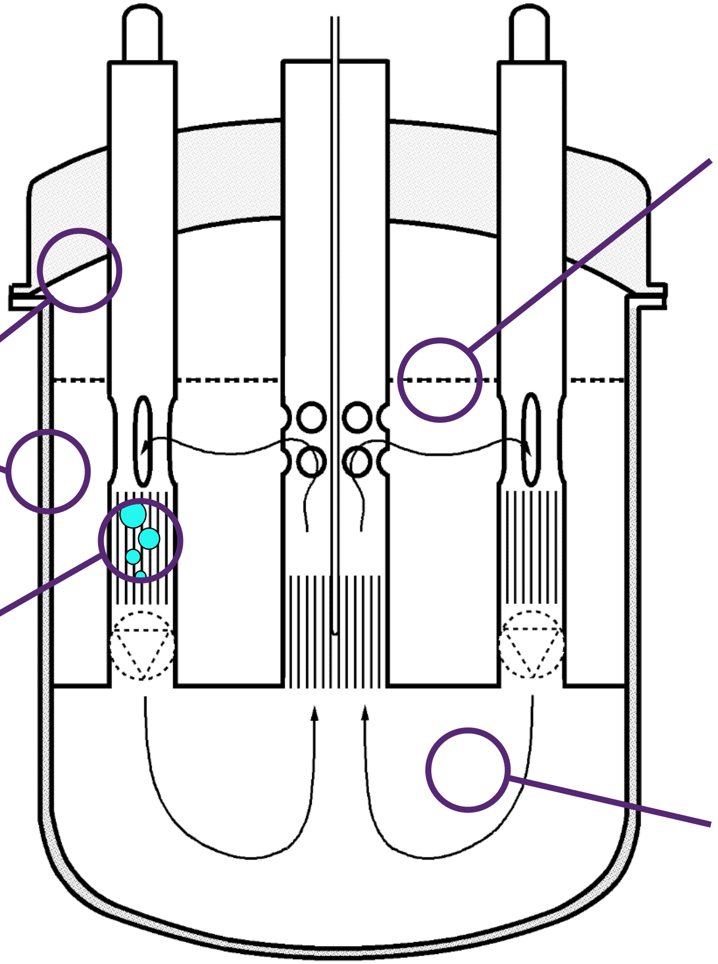
3 Fission products

Ingress of fission products from failed fuel elements (Cs, I, Te)

Radioactive impurity sources chemical reactions/phase transformation

deposition on surfaces from liquid or **adsorption** from gas

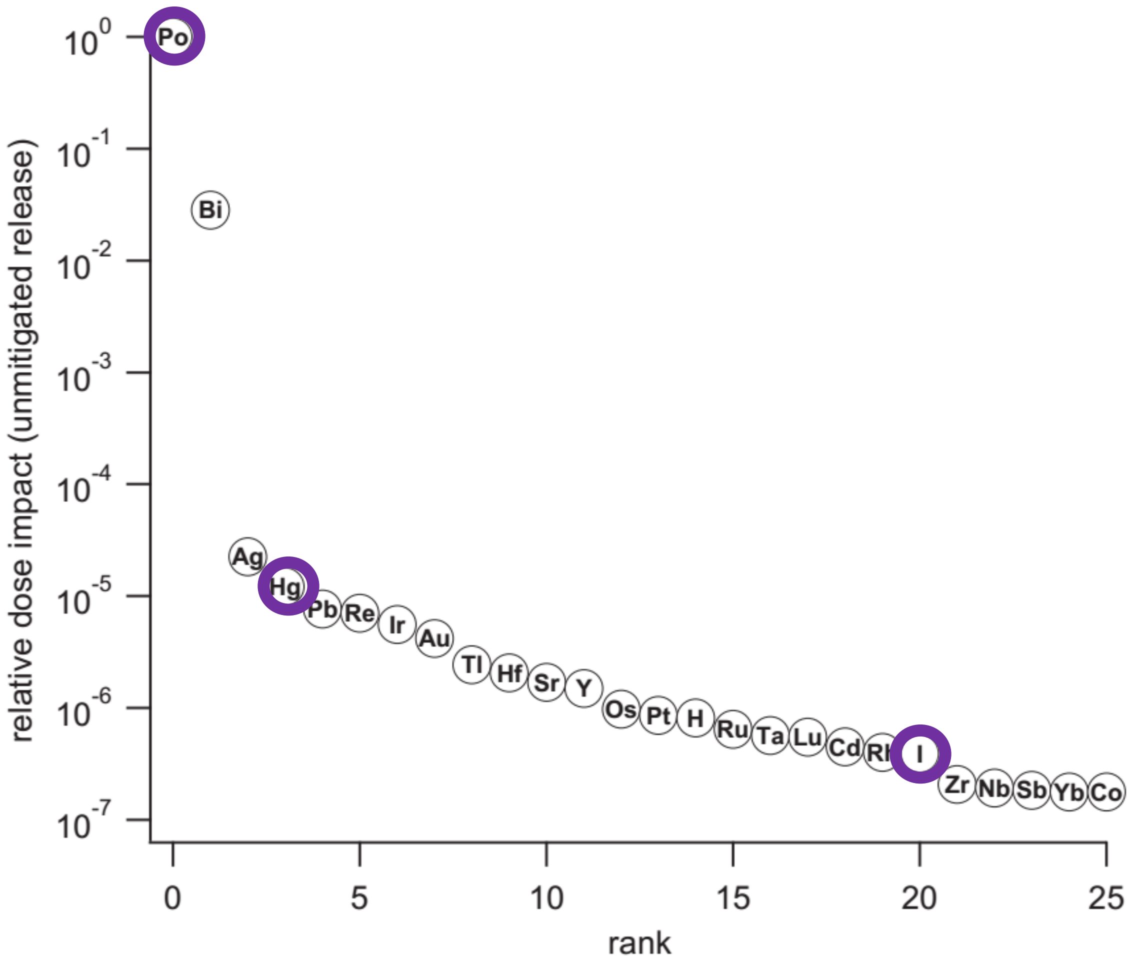
reactions with H_2O after e.g. HEX leak



- **evaporation** to cover gas
- reactions with cover gas impurities (H_2O, H_2, O_2)

- reactions with LBE, dissolved O and other impurities
- **precipitation/dissolution**

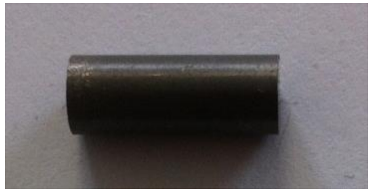
Relative dose impact of radionuclides formed in LBE



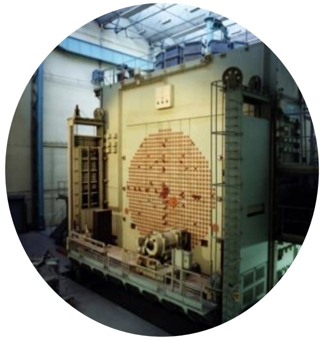
- 1st assumption: unmitigated release
- **Polonium-210 dominant**
- Chemical interactions with coolant, cover gas and structural materials strongly influence volatility/release
- R&D focus on those radionuclides combining high potential dose impact and volatility
- Knowledge of the release of radionuclides from LBE required for design and licensing

Radioactive impurity release: approach

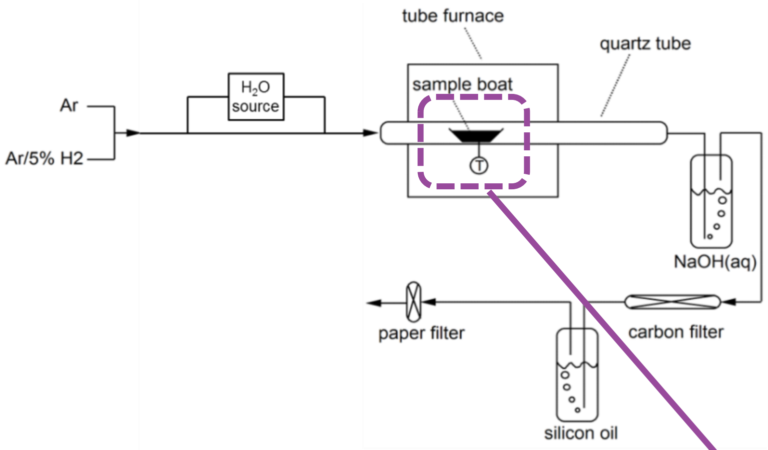
LBE



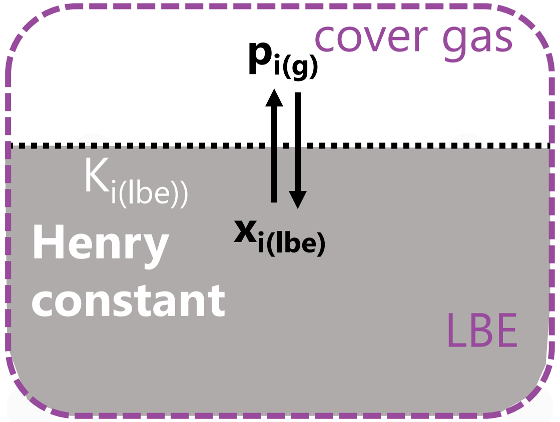
doping or irradiation in BR1 reactor



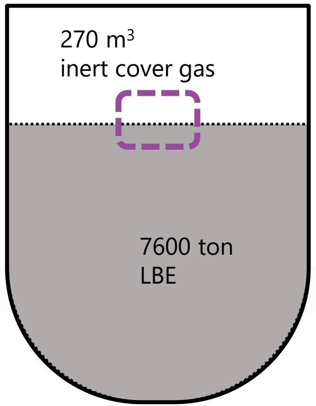
evaporation experiments



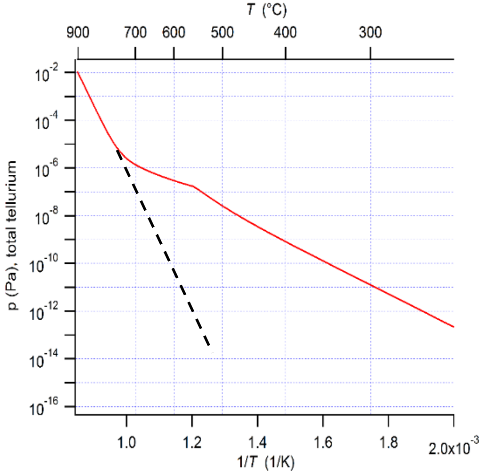
Measurement of release

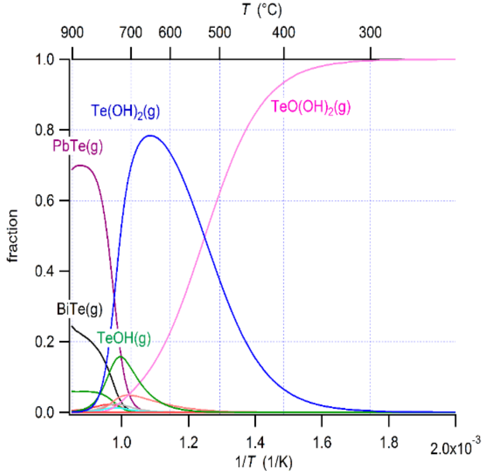


Release predictions



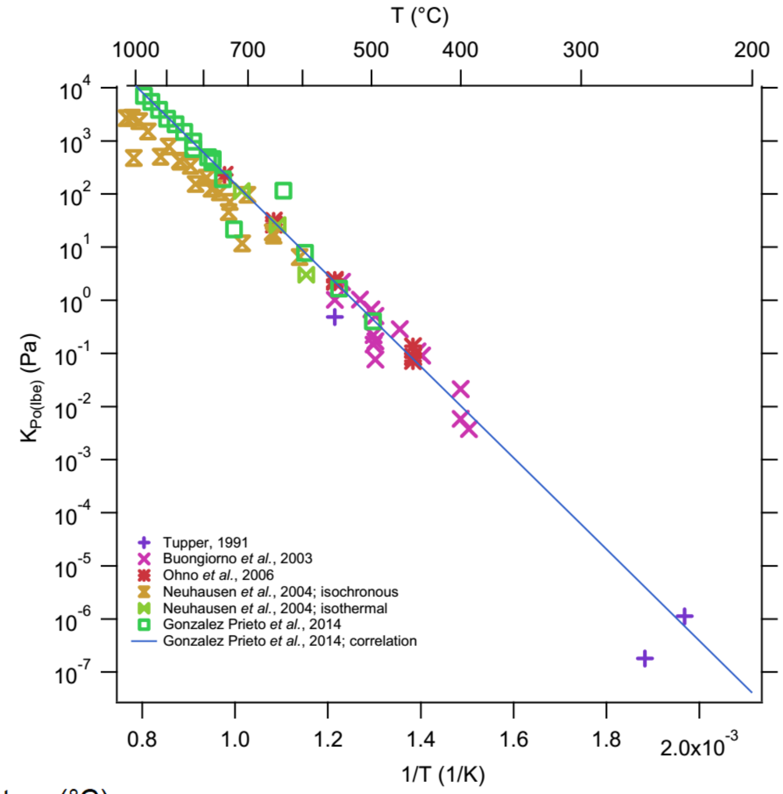
data + models

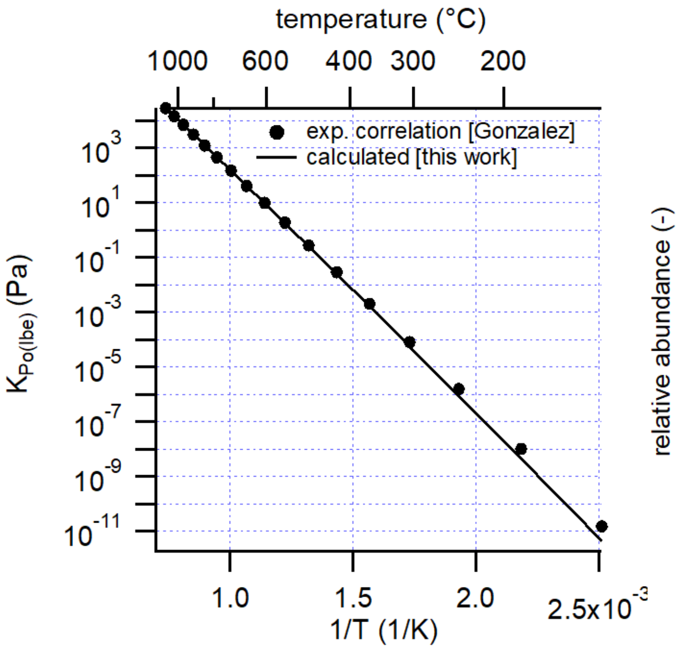


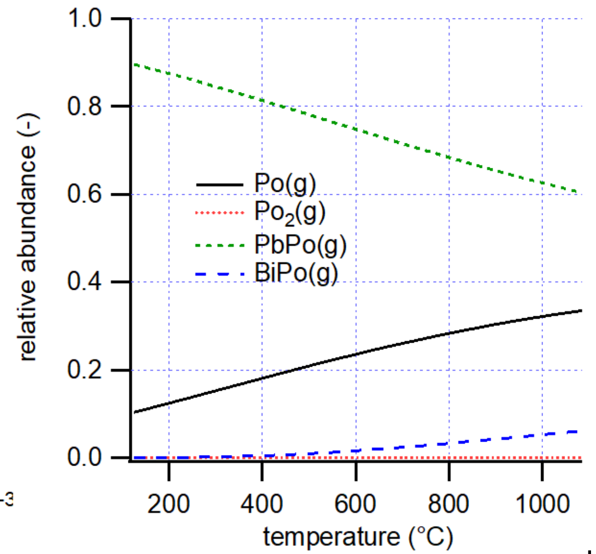


Polonium release > 500 °C

- Polonium concentration in LBE
  - Mass of ^{210}Po ≈ 2000 g → $x_{Po(LBE)}$ ≈ 10^{-7}
- Polonium is dissolved in LBE; very limited release
- Not sensitive to gas phase composition (inert, reducing, humid)
- No oxide layer (high oxygen solubility)
- Thermochemical model in agreement with experiments
- PbPo(g) and Po(g) most predominant species



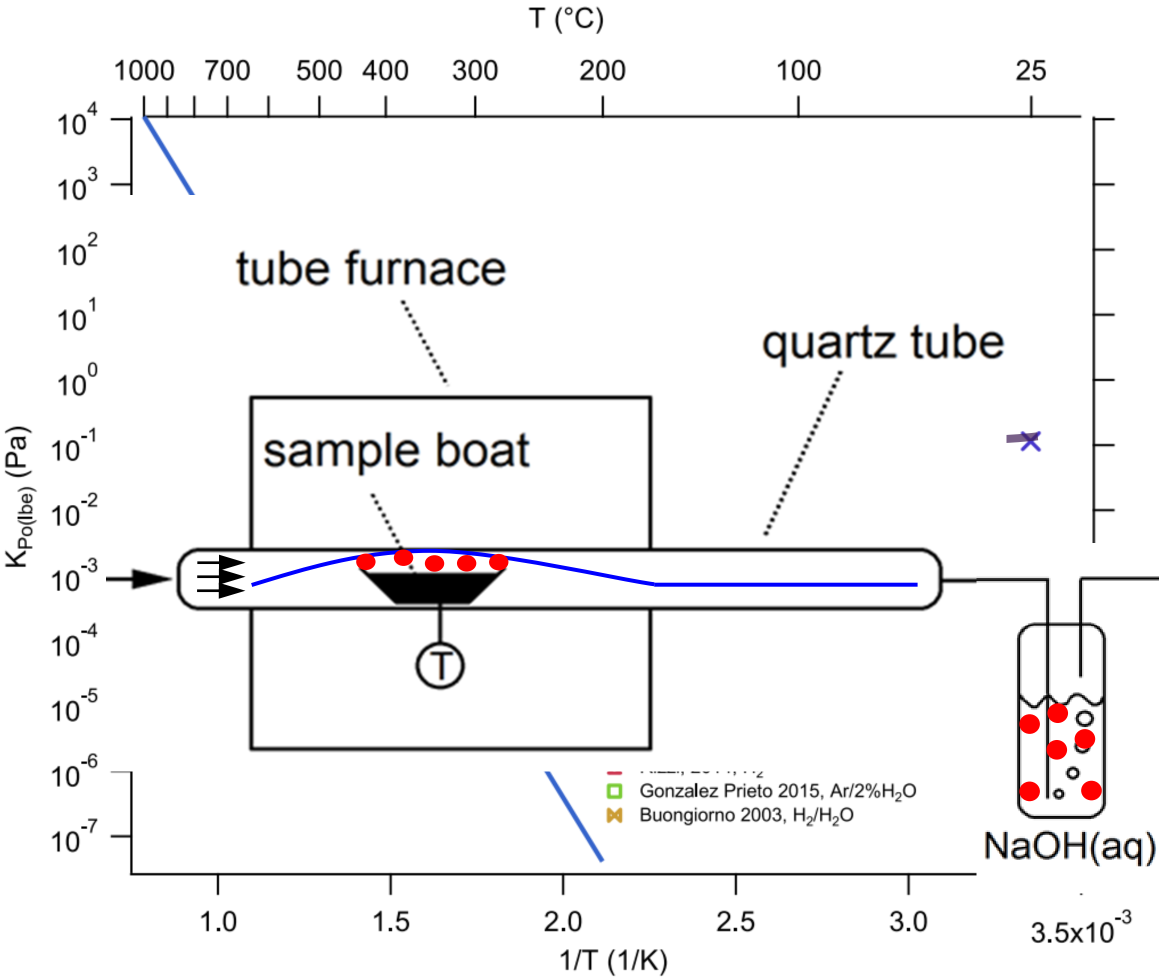




B. Gonzalez Prieto, et al. Radiochim. Acta. 102 (2014) 1083–1091.
M.A.J. Mertens, et al. J. Phys. Chem. Lett. 10 (2019) 2879–2884.
A. Aerts. *Thermo* 2021, 1–10.

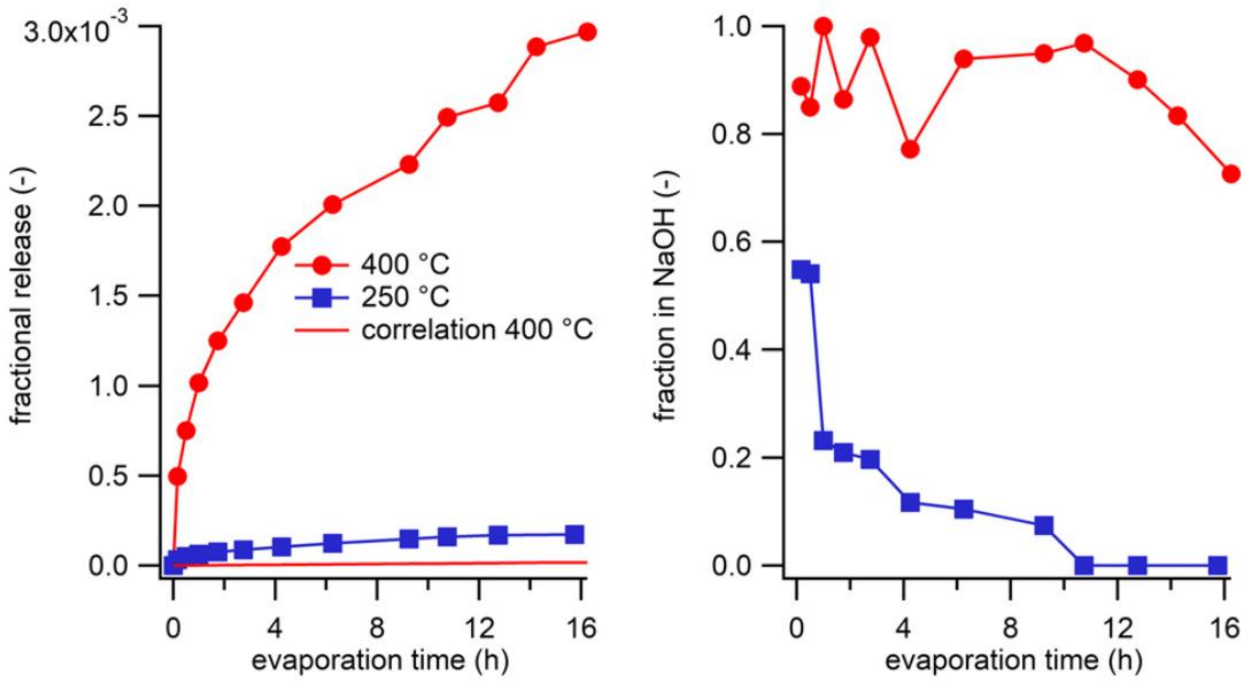
Polonium release < 500 °C

- Oxide layer on the LBE surface
- Enhanced polonium evaporation
- Release affected by surface effect (segregation)
- Release and volatility influenced by cover gas composition ($H_2O(g)$, $O_2(g)$, $H_2(g)$)



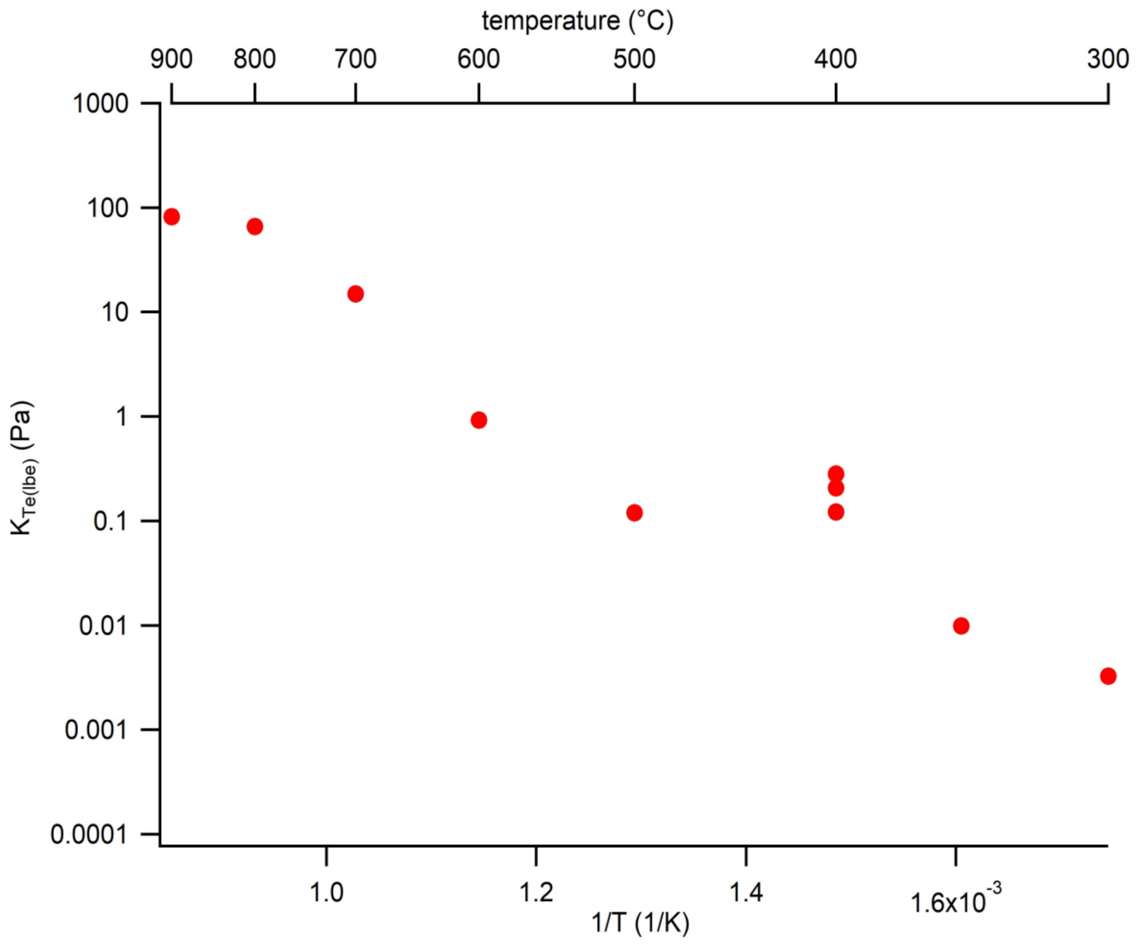
Polonium release < 500 °C: cover gas composition

- Enhanced polonium release and volatility with **H_2O vapor** and **O_2(g)**
- Interaction of Po with water vapor and O_2(g) crucial for accident analysis
- **<u>Impact on design</u>**: double walled PHX for MYRRHA



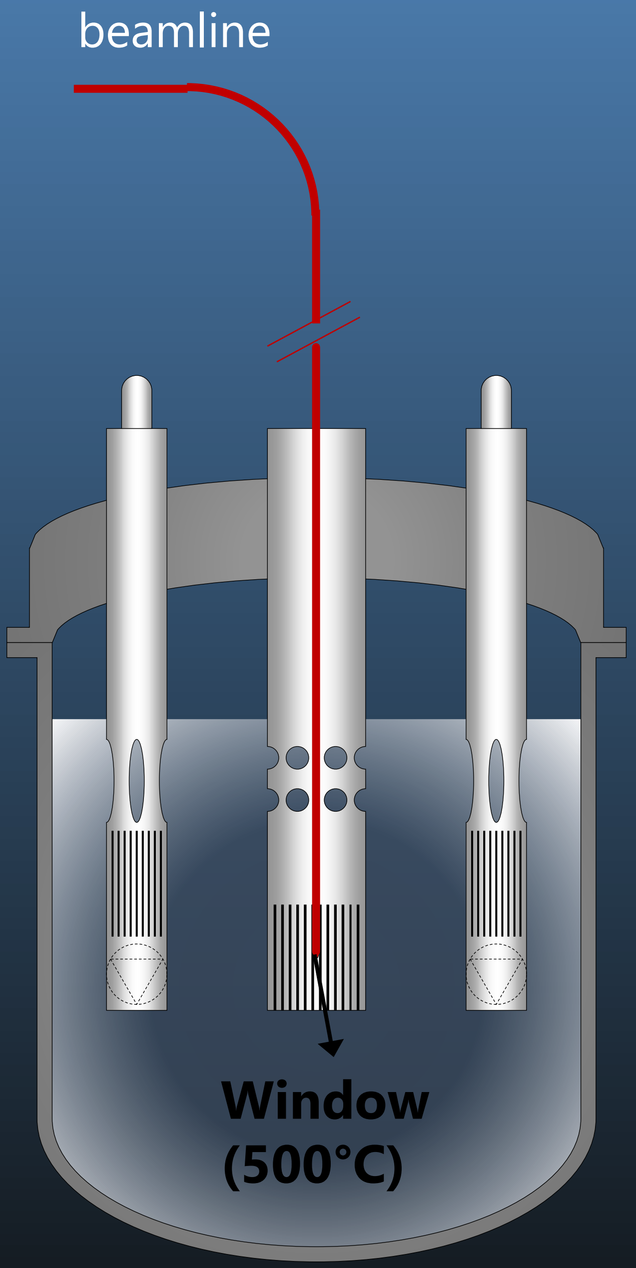
Tellurium release

- Tellurium fission product and chemical homologue of Po
- Henry-type evaporation behavior with strong retention in LBE
- Deviation from the high-temperature at T< 400 °C
- Thermochemical model reproduces the experimental results
- Enhanced released might be caused by various species formed at lower T



MYRRHA Beam tube rupture

possible source of Polonium release



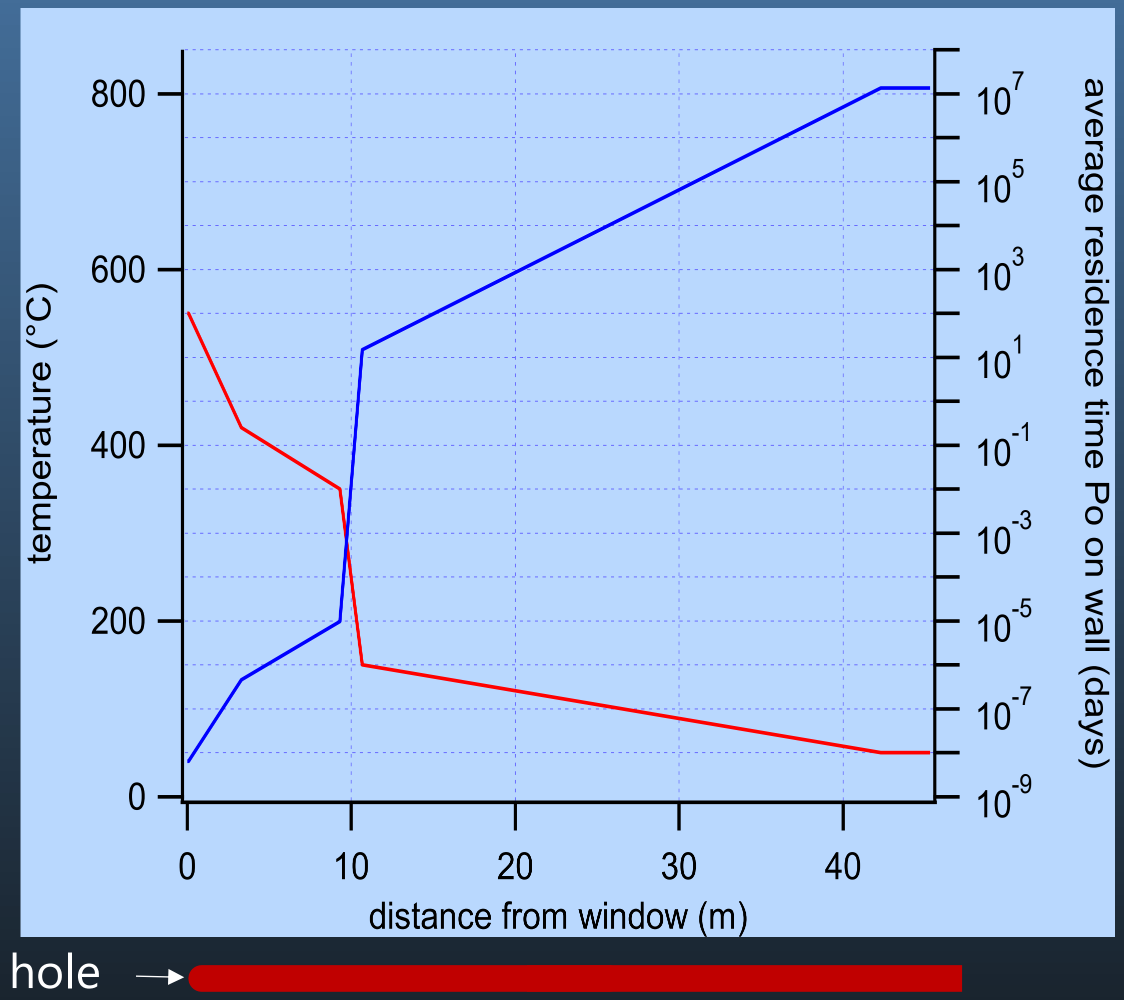
Accident scenario:

- Po released through hole in beam tube

→ *Where does it end up?*

Beam tube

- High vacuum → particles do not see each other
- Low transmission
  - long thin tube
    → many collisions
    → sticking not important
  - High residence time
- **If it sticks, it sticks**



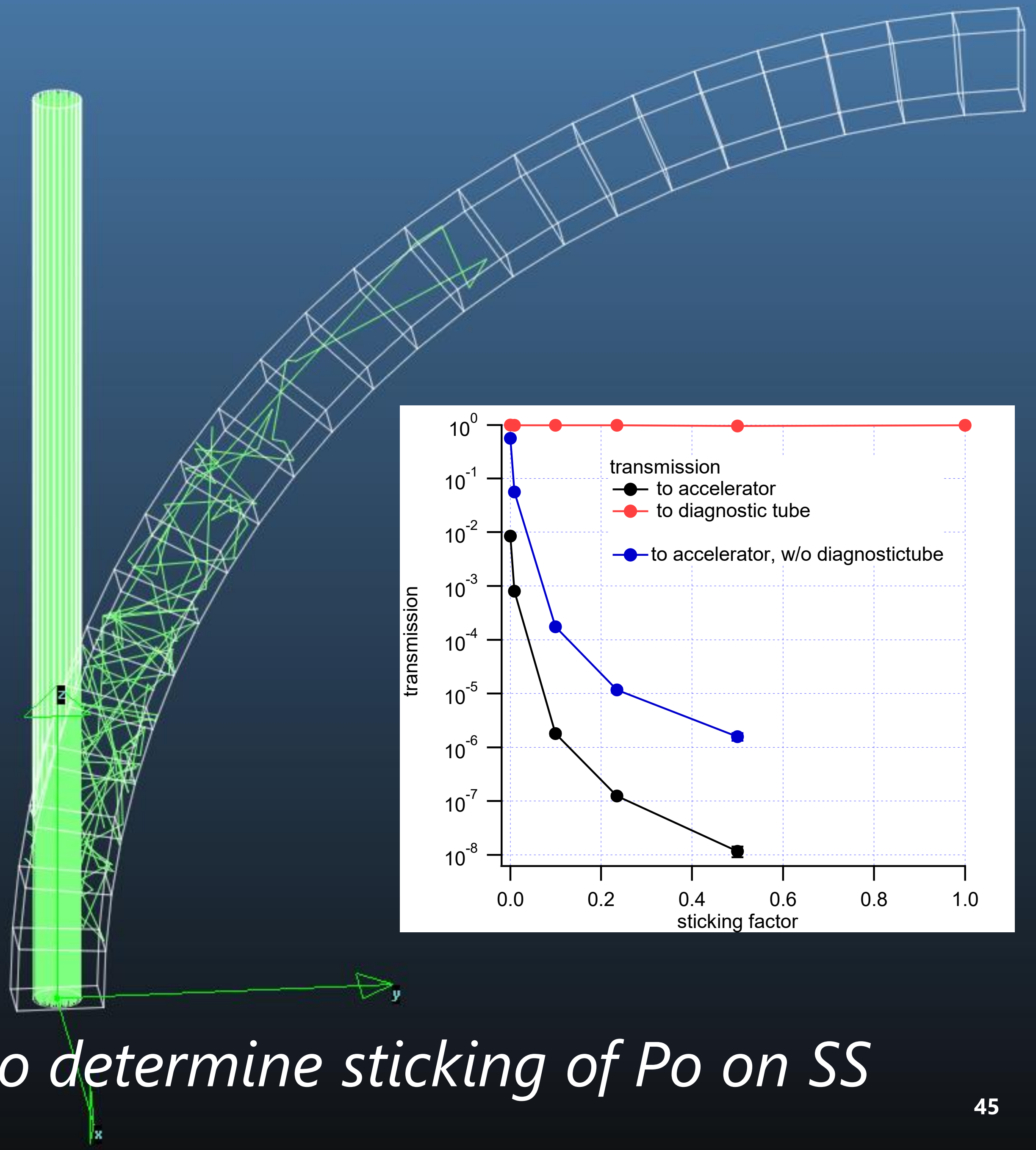
- *Beam tube transmission: ~10^{-4}*

Bending magnet

Transmission

- Beamtube * bend
- Beam tube: ~10^{-4}
- bend: sticking factor dependent

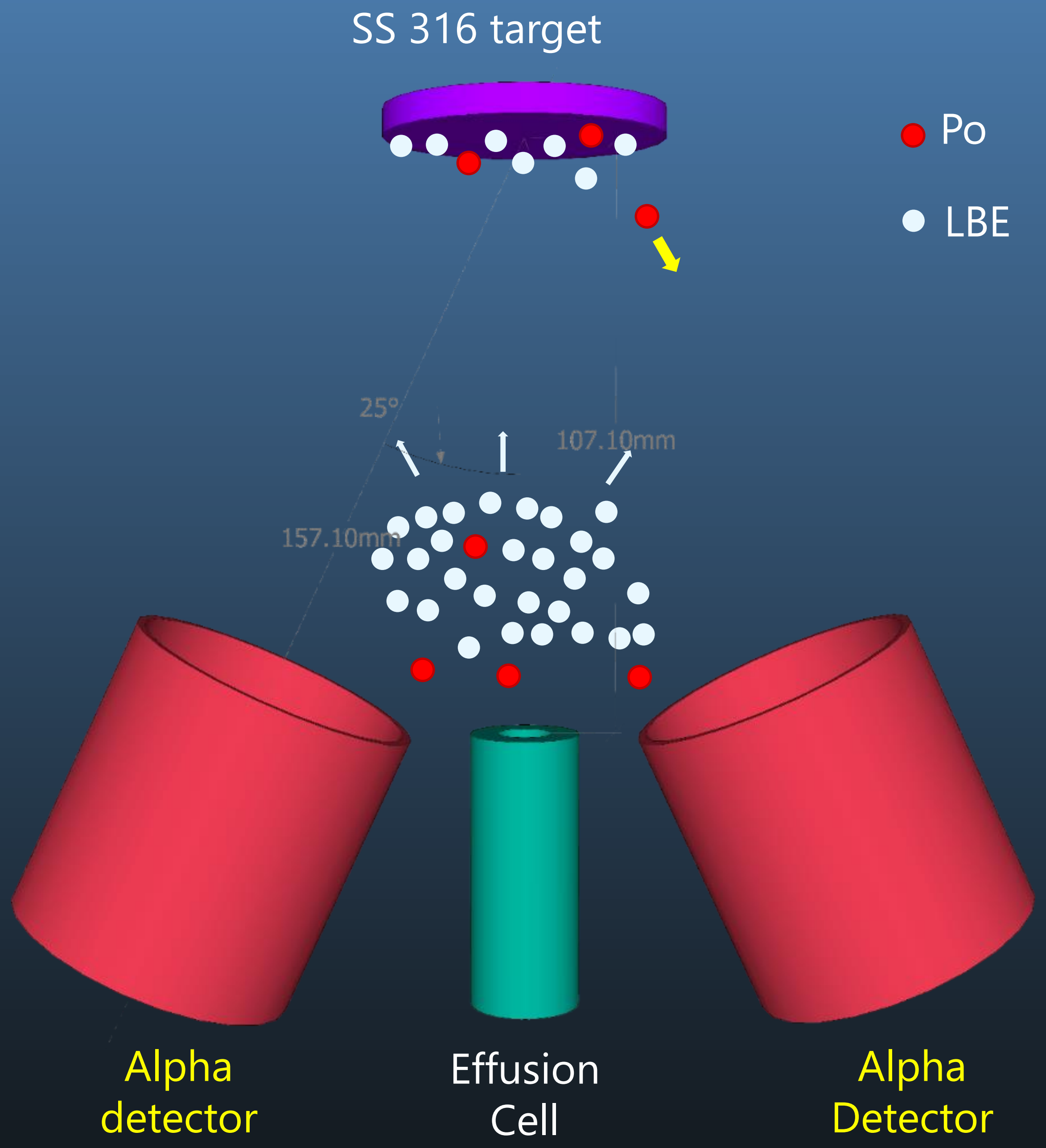
Total transmitted: ~10^{-4} to 10^{-10}



→ *We need to determine sticking of Po on SS*

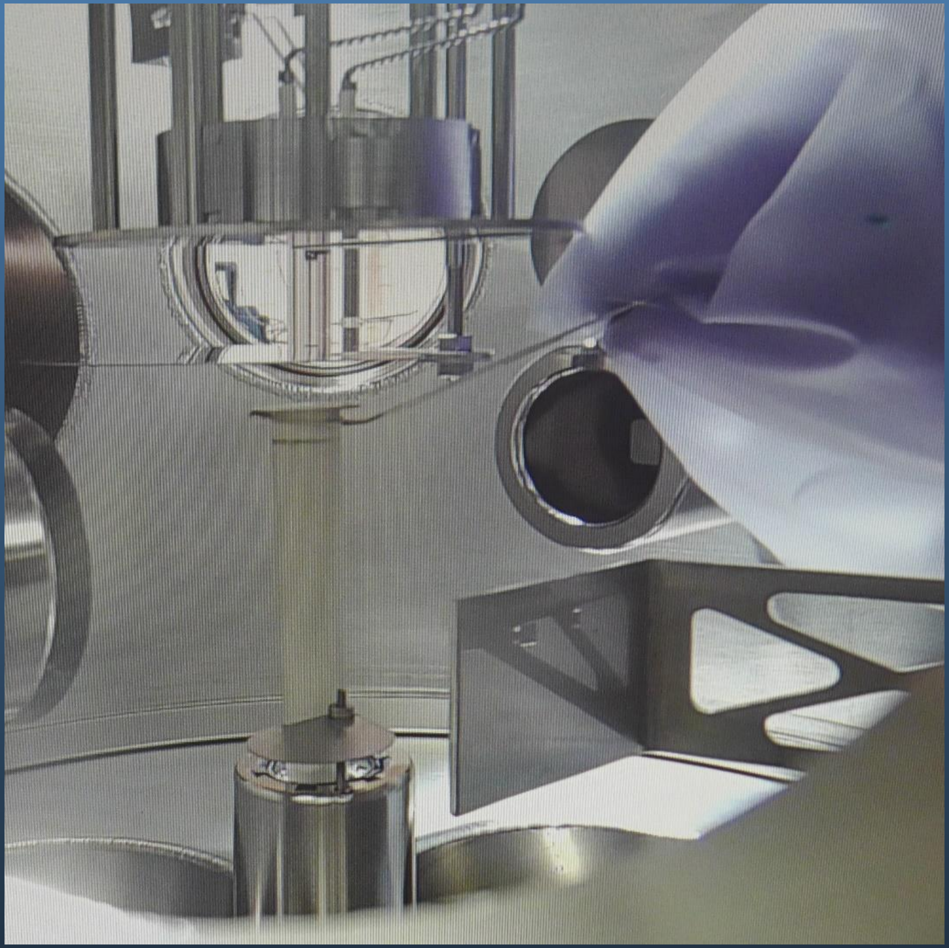
Principle

- Evaporate LBE + Po
- Po fractions?
  - Eff cell → target: f_1
  - Target → alpha: f_2 (s)
- Sticking:
  - Target: s=?
  - Alpha: s=1
- Sticking: $f_1 * f_2$
- Compare f_1 and f_2 from simulation with experiment



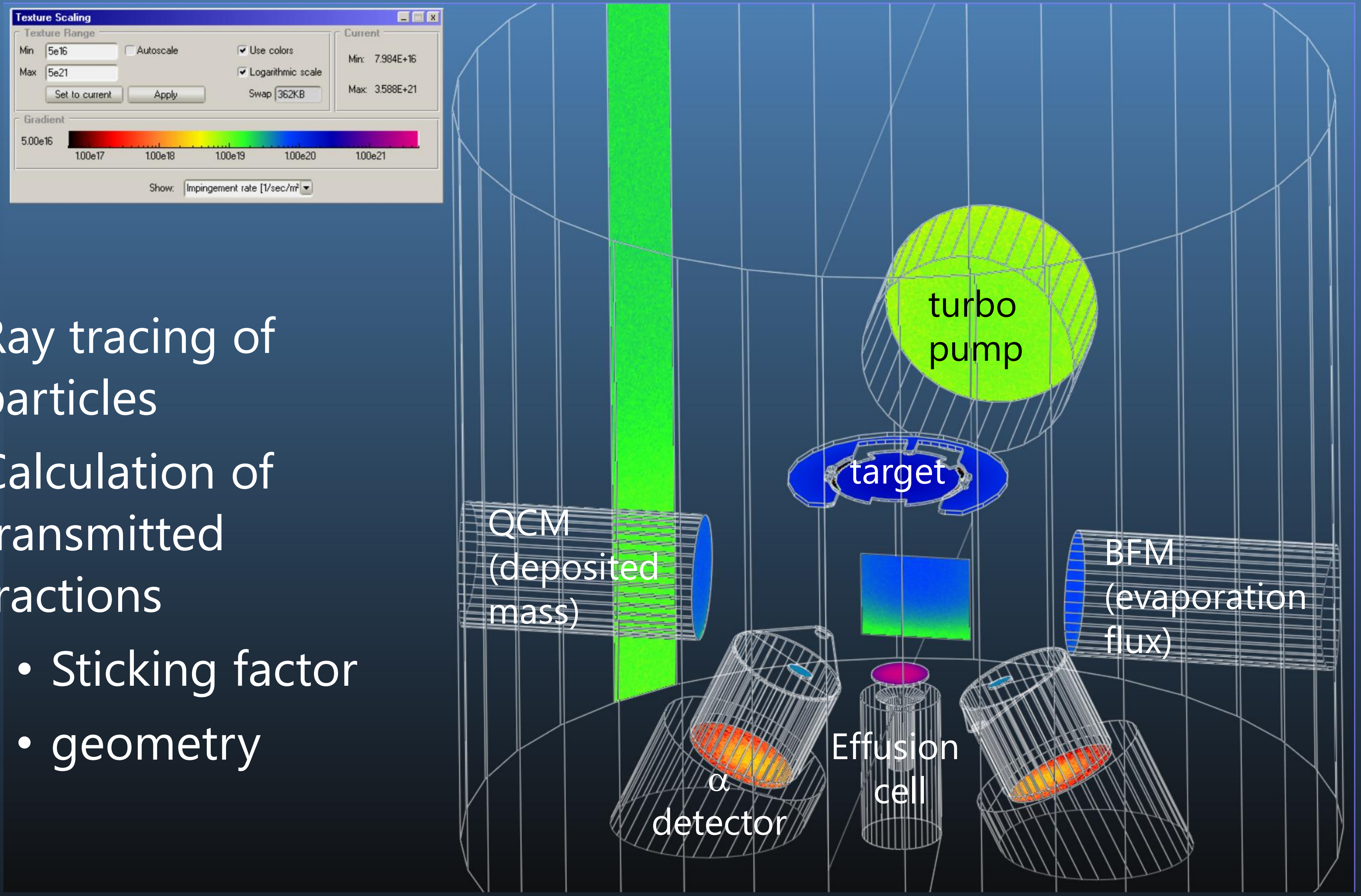
POVACS Setup

General overview



Licensing:
Avoid Po contact with the environment

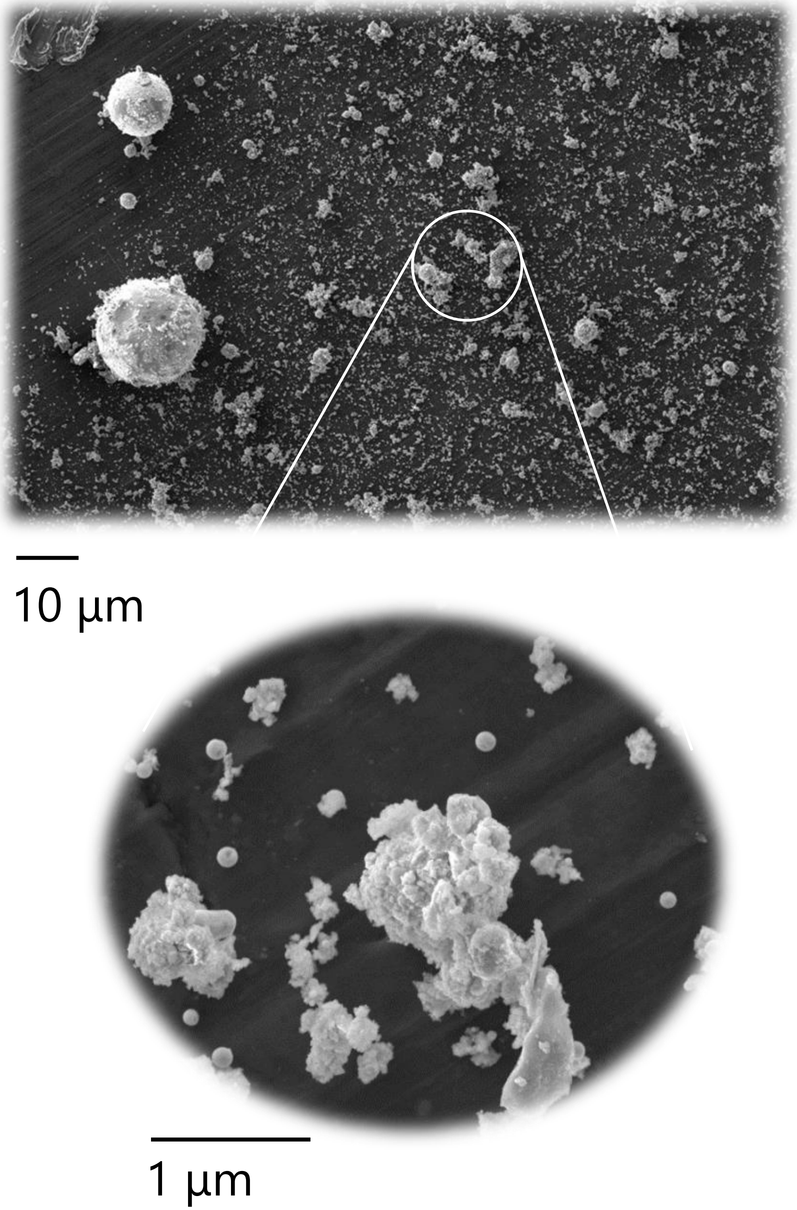
MOLFLOW simulations

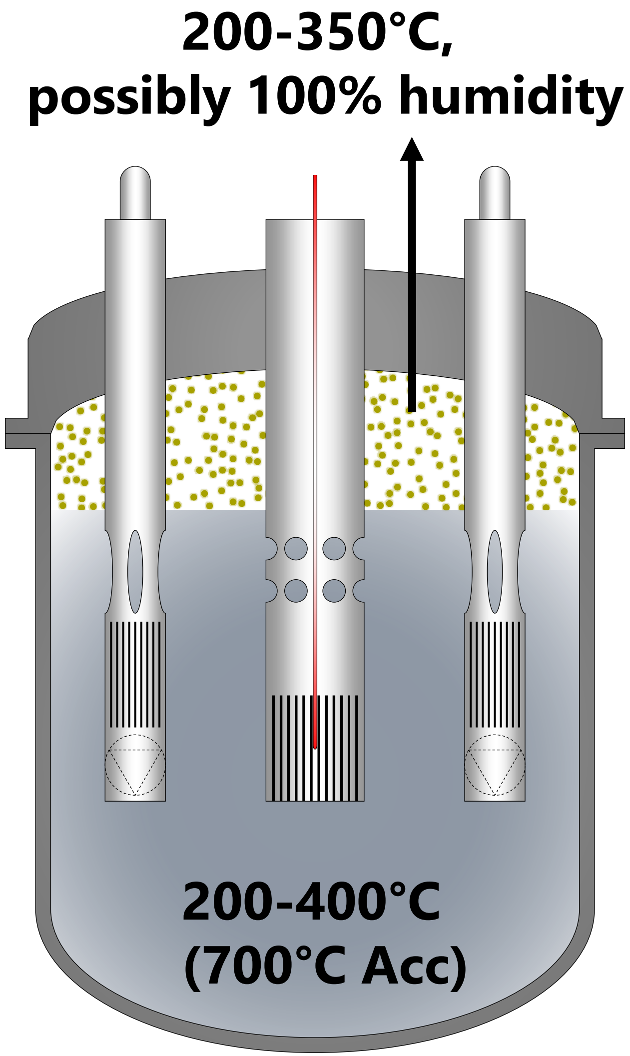


- Ray tracing of particles
- Calculation of transmitted fractions
    - Sticking factor
    - geometry

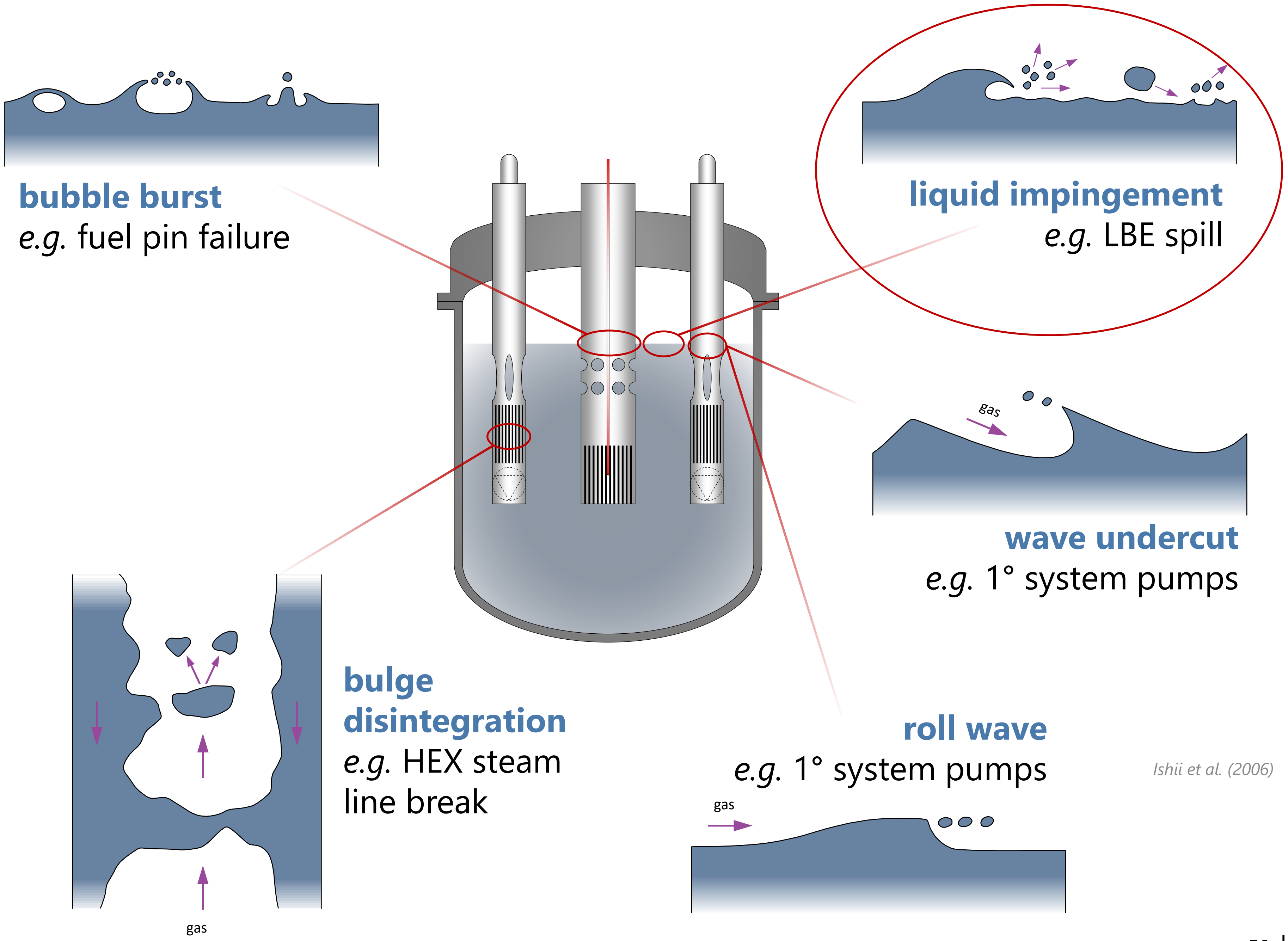
Aerosols in MYRRHA

Potential source of radioactive release



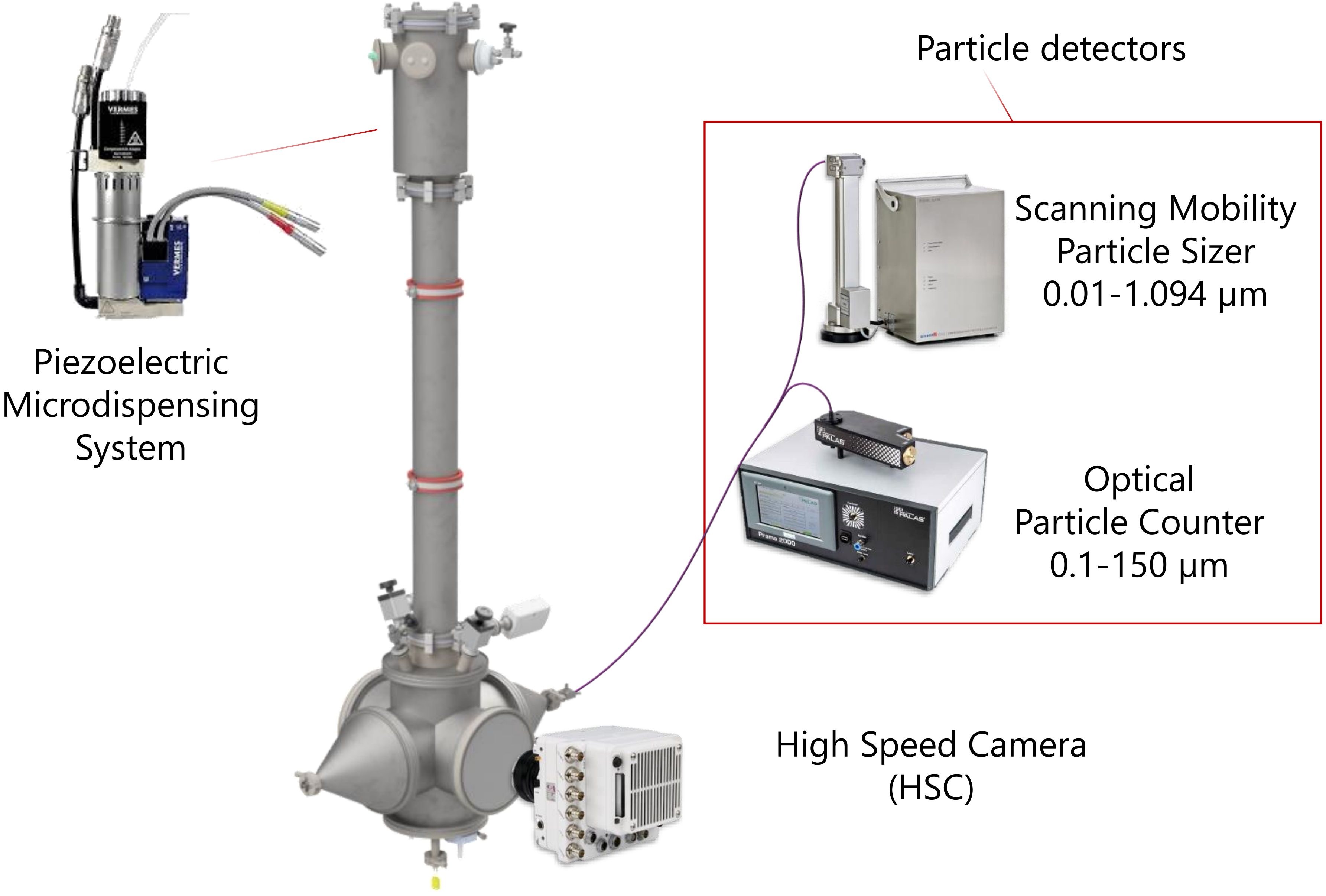


Aerosol formation



Ishii et al. (2006)

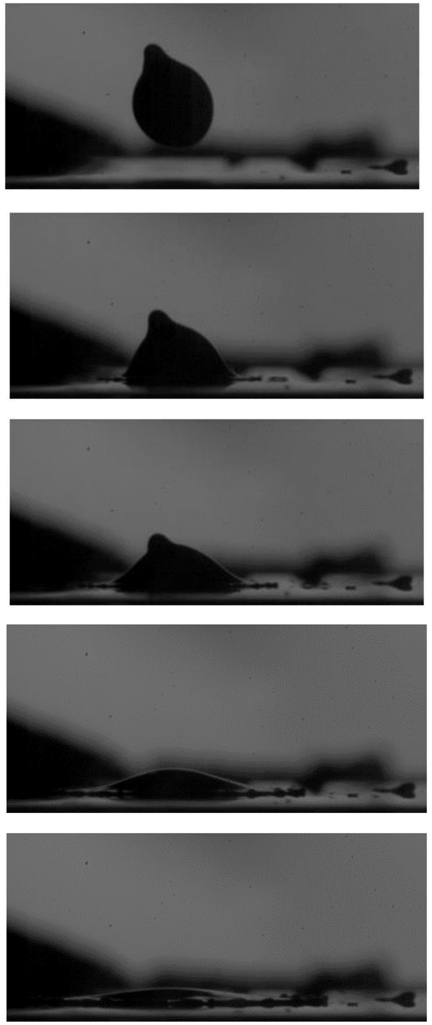
The SPLAT setup



Splashing of LBE droplets

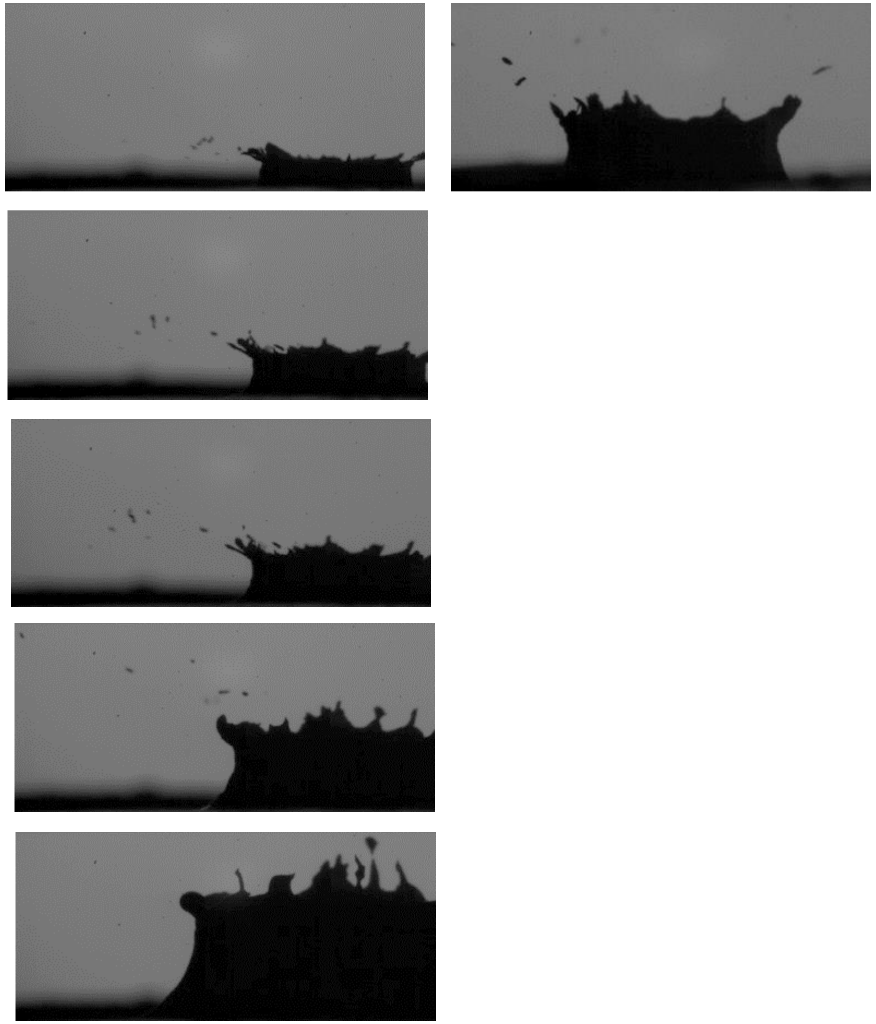
Solid impingement

Prompt splash



Liquid impingement

Crown splash



Key messages to remember

Concluding remarks

- LBE is fundamentally very similar to pure Pb
  - Thermal-hydraulic differences are negligible
  - Focus on coolant chemistry
- Chemistry kinetics at low temperatures (~200°C) is slower
- Corrosion products: Bi can lead to formation of Bi_3Ni crystals
- Bi leads to formation of large amounts of ^{210}Po as an activation products
- Pure Pb chemistry is simpler to control
- Main advantage of LBE is lower melting point → ADS applications

Thank you!

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