26th International Conference on Nuclear Engineering (ICONE26)
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PRACTICAL EXPERIENCE GAINED FROM OPERATING THE LEAD-BISMUTH LOOP CORRIDA

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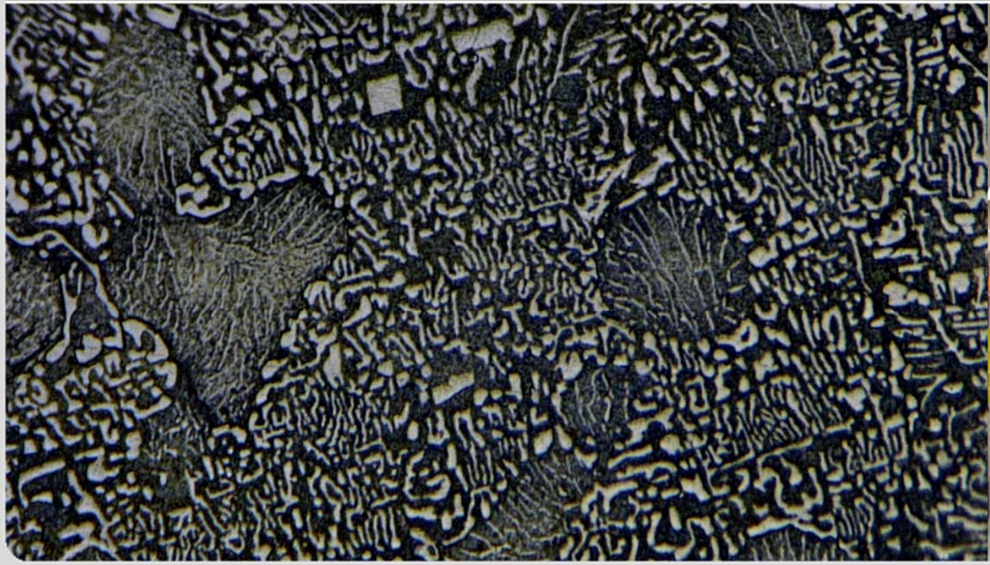
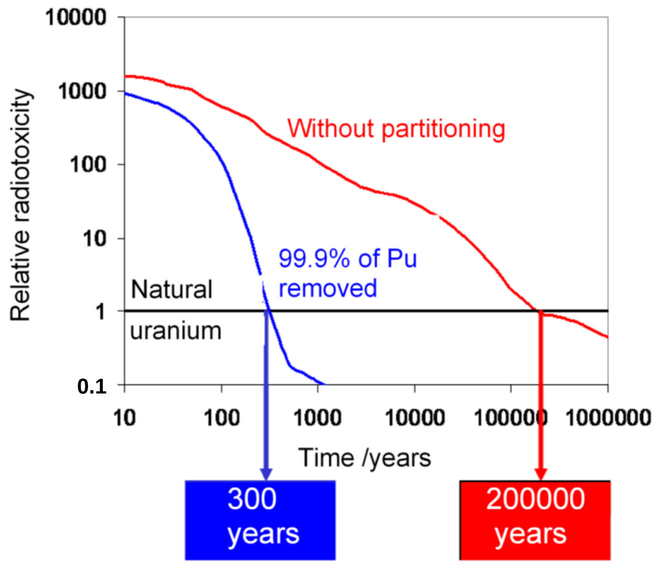
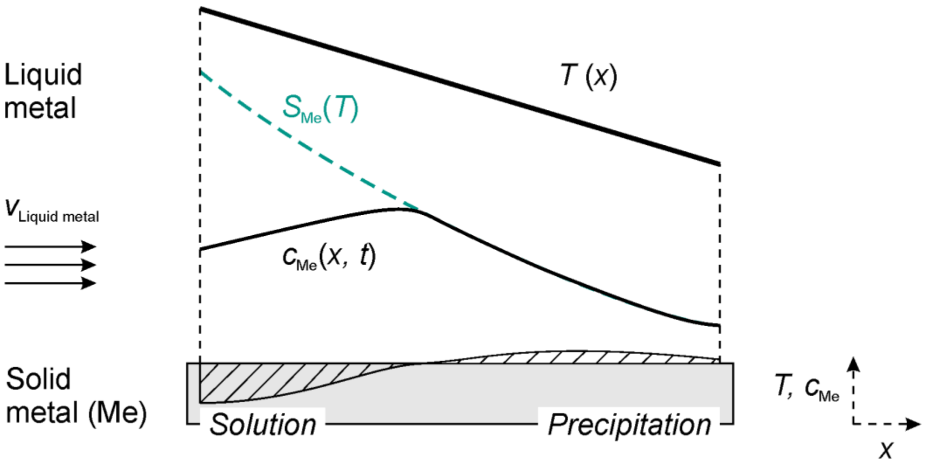


Background

- Renewed interest in liquid lead and lead-bismuth eutectic (LBE) as coolants for nuclear reactors.
  - Lead-cooled fast reactor is one of the concepts selected for the fourth generation of nuclear power plants.
  - LBE is a candidate coolant and spallation-neutron source for the transmutation of long-lived isotopes in an accelerator driven sub-critical system (ADS).



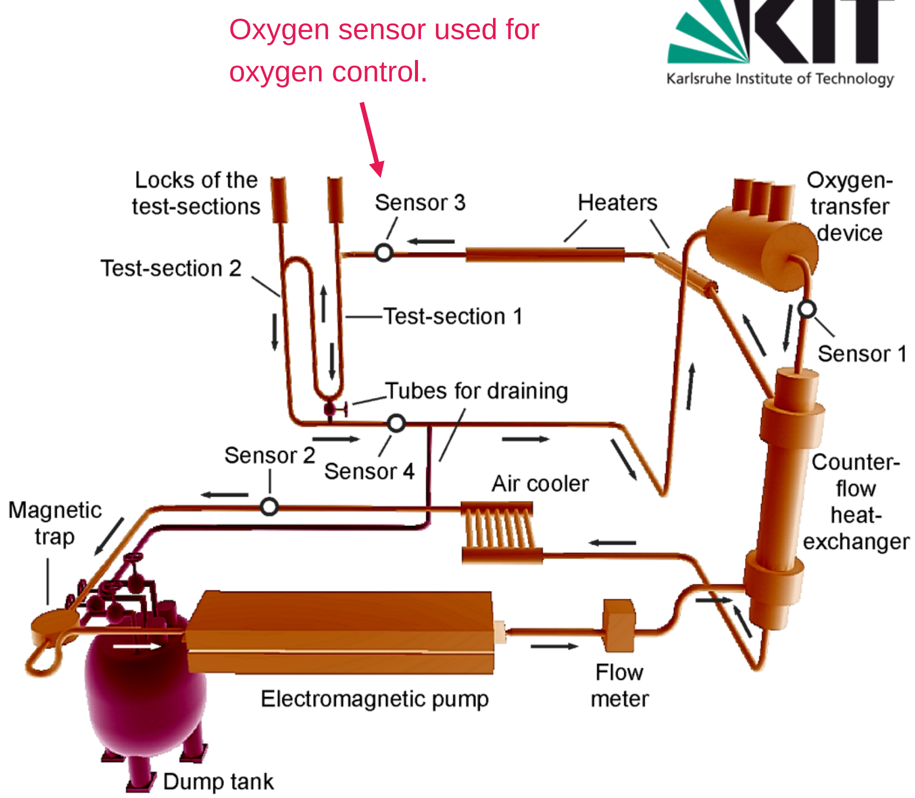
Benefit of partitioning and transmutation of nuclear waste as to final storage of the residues.



Solution and re-precipitation of solid metal determined by the temperature-dependent solubility S_{Me}.

- Particular challenges identified in the very early studies:
  - Compatibility with steels.
  - In non-isothermal system, plugging of the cold leg in consequence of material corrosion in the hot part.
  - Gain from oxygen addition to lead or LBE?

About the CORRIDA loop



- Important technical characteristics:
  - LBE inventory of 2500 kg.
  - 1000 kg LBE circulate in the loop, driven by electromagnetic pump.
  - Mass flow typically 5.3 kg/ s.
  - Up to 550 °C in the hot sections at a minimum temperature of ~300–385 °C in the cold leg.
  - Tubing and main components made of Steel 1.4571 (~316Ti).
  - Gas/ liquid oxygen transfer.
  - Oxygen transfer in the hot leg but on the way back to the cold section.

- Main aim:
  - Long-term corrosion tests on steels in flowing lead-bismuth eutectic at 2 m/ s flow velocity, temperature between 400 and 550 °C, and 10^{-7} to 10^{-6} % dissolved oxygen.

- "By-products":
  - Performance of the loop itself during long-term operation.
  - Fundamental aspects of gas/ liquid oxygen transfer.
  - Long-term performance of electrochemical oxygen sensors.

Details of oxygen transfer

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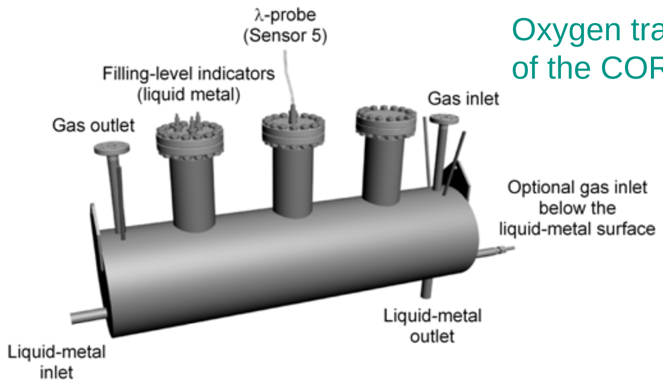
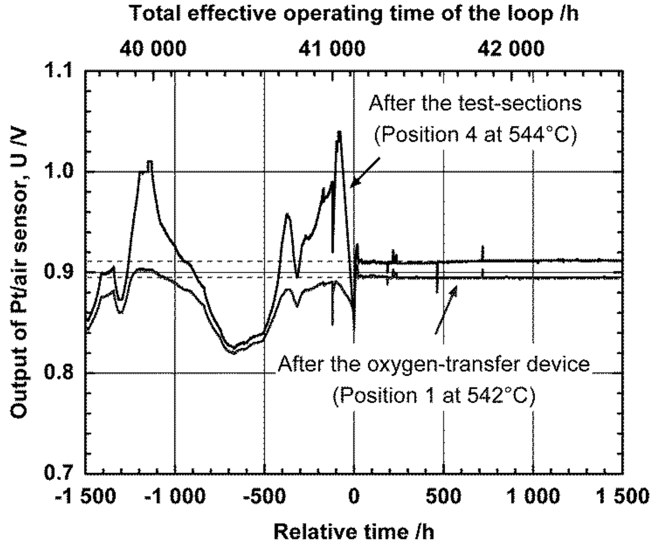
- Gas/ liquid oxygen transfer
  - Across a plane LBE surface (~0.3 m²).
  - Gas introduced above the liquid-metal surface.
  - Quasi-static gas, slow-flowing liquid metal (0.025 m/ s at 5.3 kg/ s).
  - Commercial λ-probe monitoring the oxygen partial pressure in the gas plenum.



Oxygen transfer device of the CORRIDA loop.

- Gas supply
  - Up to 500 Nccm/ min argon.
  - Up to 500 Nccm/ min argon–hydrogen (5 % H_2) for regular operation, 2000 Nccm/ min for pre-conditioning.
  - Up to 10 Nccm/ min air.
  - Humidifying the gas was abandoned with the automation of air addition.



Before and after implementation of automated air addition.

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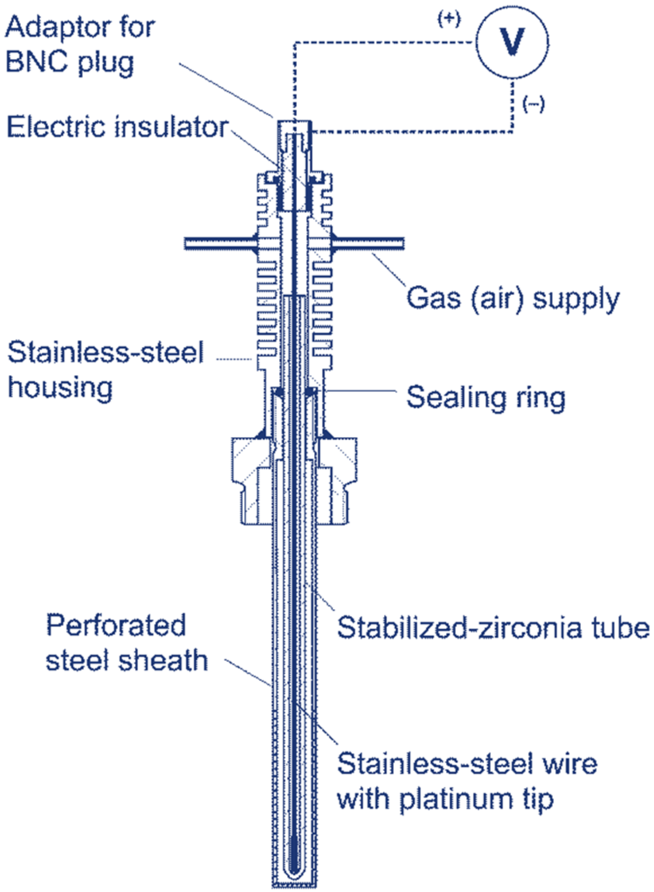
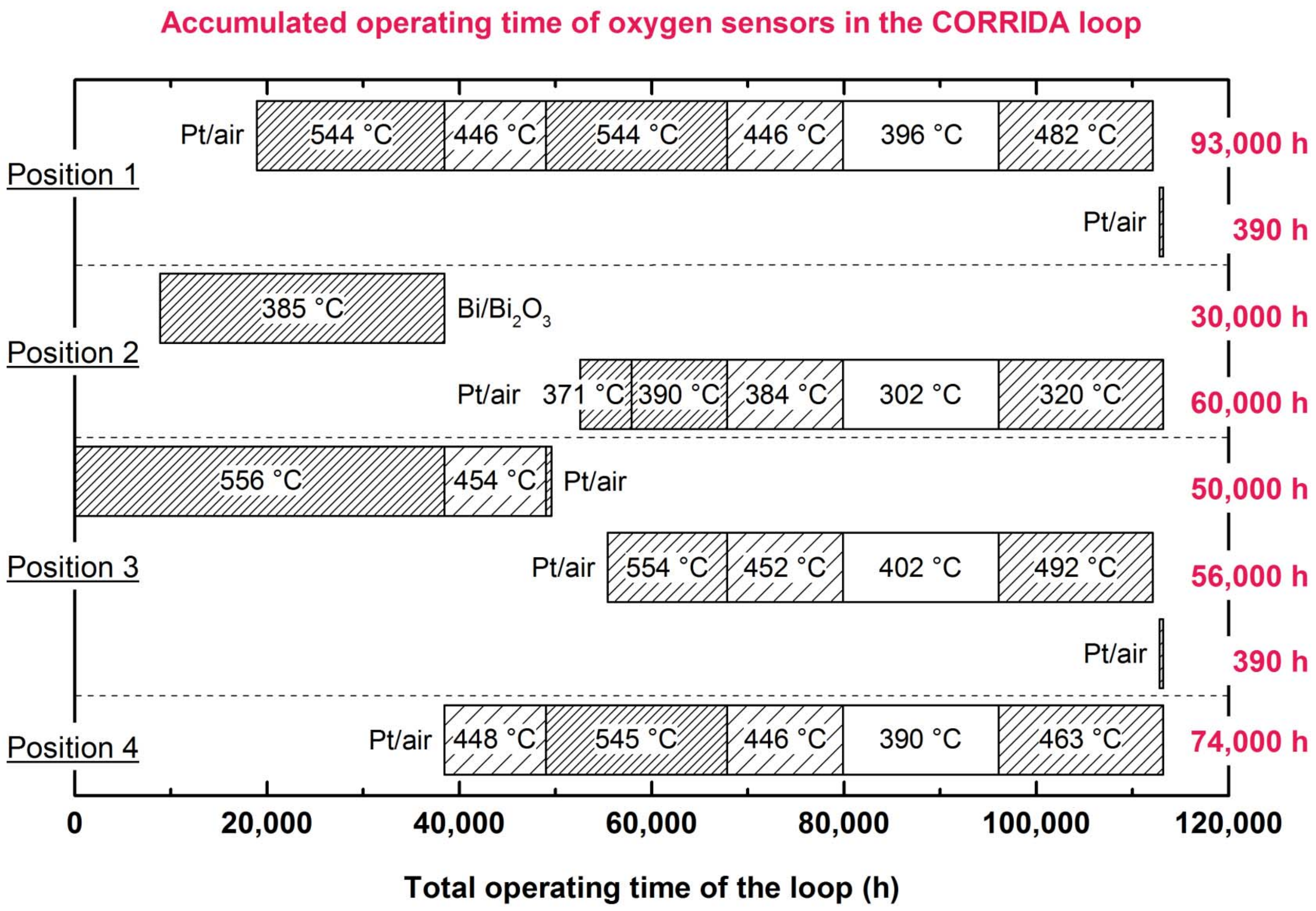
Electrochemical oxygen sensors



Design of oxygen sensors installed to the CORRIDA loop.

- Pt/ air or Bi/ Bi_2O_3 reference electrode, with preference of Pt/ air.
- Electrolyte: γ-zirconia partially stabilized with yttria.
- Long electrolyte tube housed in a perforated steel sheath.
- Accuracy of +/– 5 mV or better in tests against the Pb/ PbO equlibrium at 350–650 °C (Pt/ air).
- Four sensors in selected positions along the CORRIDA loop.

Operating time of sensors in the loop



Operating history of the CORRIDA loop

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Commissioning in Feb 2003.

Start of operation at 550 °C/ 10^{-6} % oxygen in Jul 2003.
Oxygen transfer with $Ar–H_2–H_2O$.

Manual air addition to $Ar–H_2–H_2O$. ☺

Optimization.

Dry gas.

Optimized manual air addition to humidified Ar. ☺
Tube failure at the inlet of the second test-section after total operation for 29,000 h. ☹

450 °C/10^{-6} % oxygen.

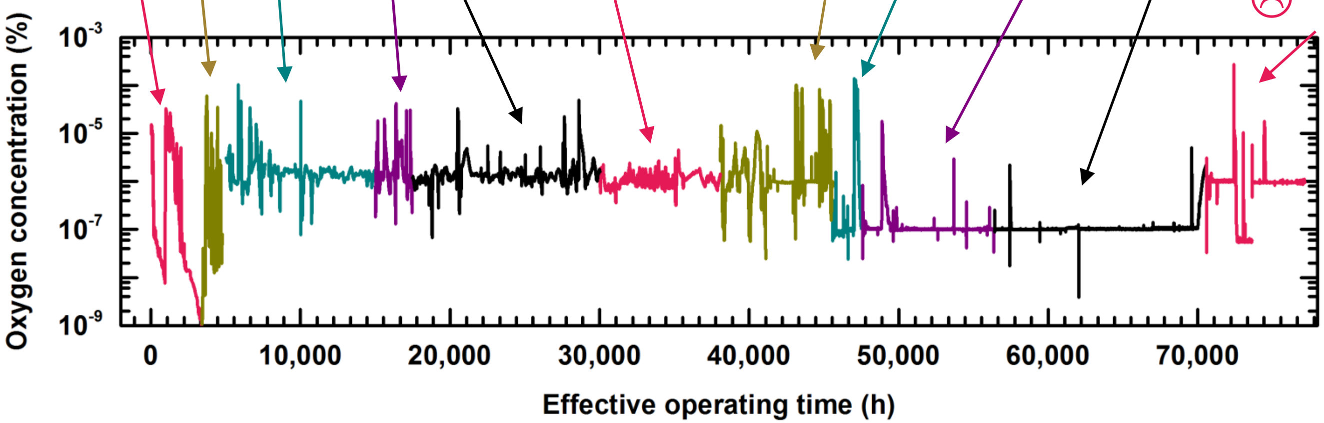
Return to 550 °C/ 10^{-6} % oxygen.
Automated air addition to Ar or $Ar–H_2$. ☺
Decreasing mass flow and mobile oxide deposits. ☹

☺ Automation of hydrogen addition.
550 °C/ 10^{-7} % oxygen.
☹ Tube failure at the bottom end of the first test-section after total operation for 66,000 h.

450 °C/ 10^{-7} % oxygen.
☹ Tube failure close to the weldment of the once replaced inlet piece after total operation for 47,500 (76,500) h.

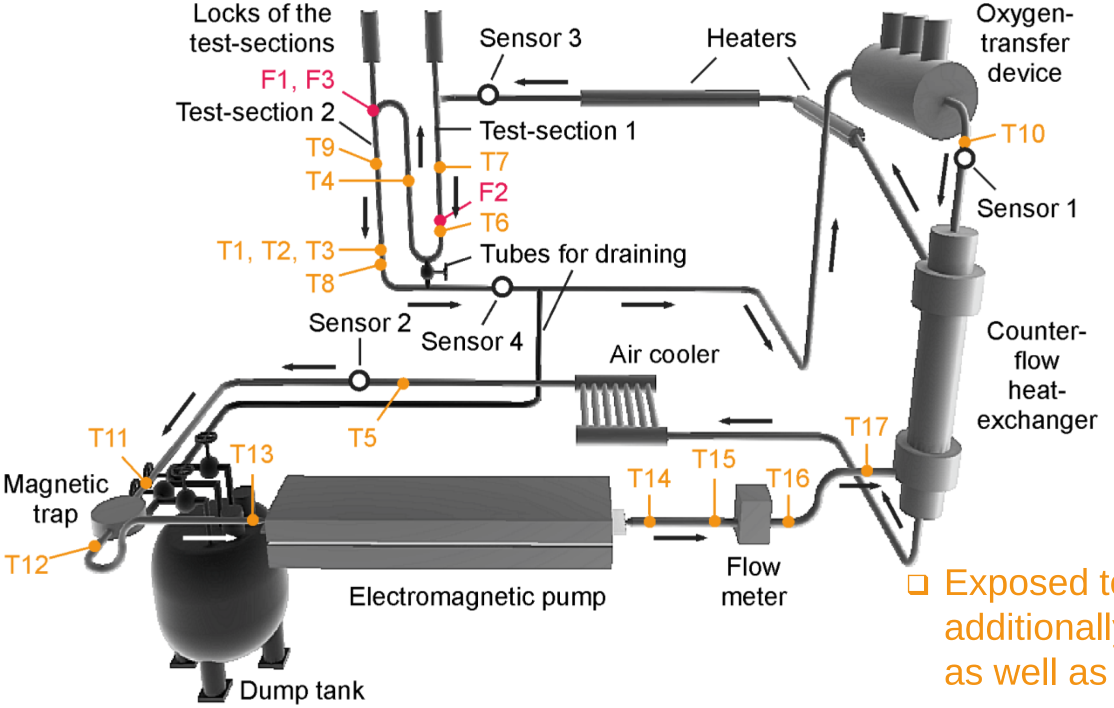
400 °C/ 10^{-7} % oxygen.

500 °C/ 10^{-6} % oxygen.
☹ Problems with mass flow return.
Total operation for 100,000 h in Feb 2016. ☺
Effective operation for 75,000 h in May 2017.



Plug in the cold leg after total operation for 113,000 h (Dec 2017): ☹
Loop cannot be refilled after LBE dump.

Probing of the tubing of the loop

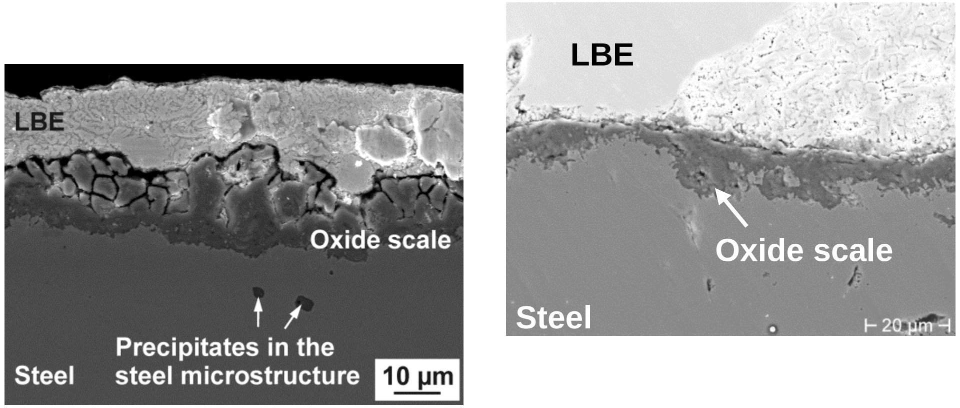


- Tube failures in the hot leg
  - Typical corrosion damage after operation of the tube for 66,000 h (F2).
  - Detrimental effect of local flow patterns suspected for F1 (29,000 h) and F3 (47,500 h).

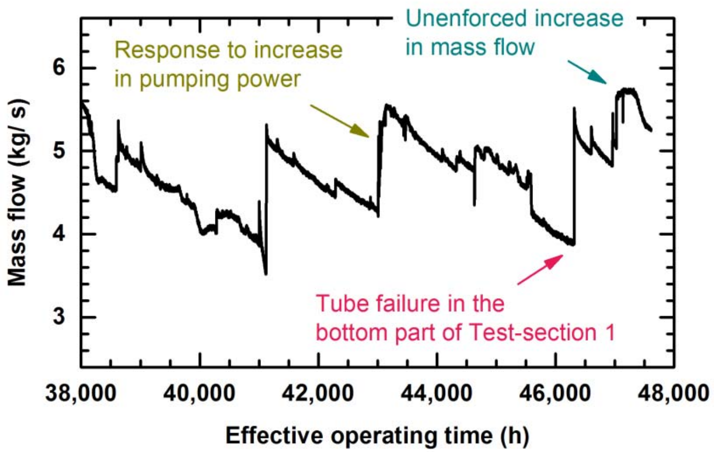
- Tube samples
  - All in all 9 from the hot leg (T1–T4, T6–T10) after operation of the tubes for 6000 (T3) to 108,000 h (T10).
  - Probing of the cold leg after 40,000 (T5) and 113,000 h (T11–T16).
  - Nominal flow velocity of 2 (T7), 1.7 (T1–T4, T6, T8, T9) or 1 m/s (T5, T10–T16).

- Exposed to flowing LBE mostly at 550 °C, additionally at 450 °C and 10^{-7} % oxygen (T6–T9) as well as 400 °C/ 10^{-7}% (T10).
- Samples from the cold leg mostly experienced 385 °C and 10^{-6} % oxygen or higher (T5), additionally 350 °C and 10^{-7} % oxygen (T11–T16).
- Pre-oxidized surface and periods of low oxygen concentration in the beginning of the exposure except for T3, T8 and T9.
- Tests on Steel 1.4571 under constant conditions available from the dedicated corrosion studies performed in the loop.

Corrosion in the hot leg



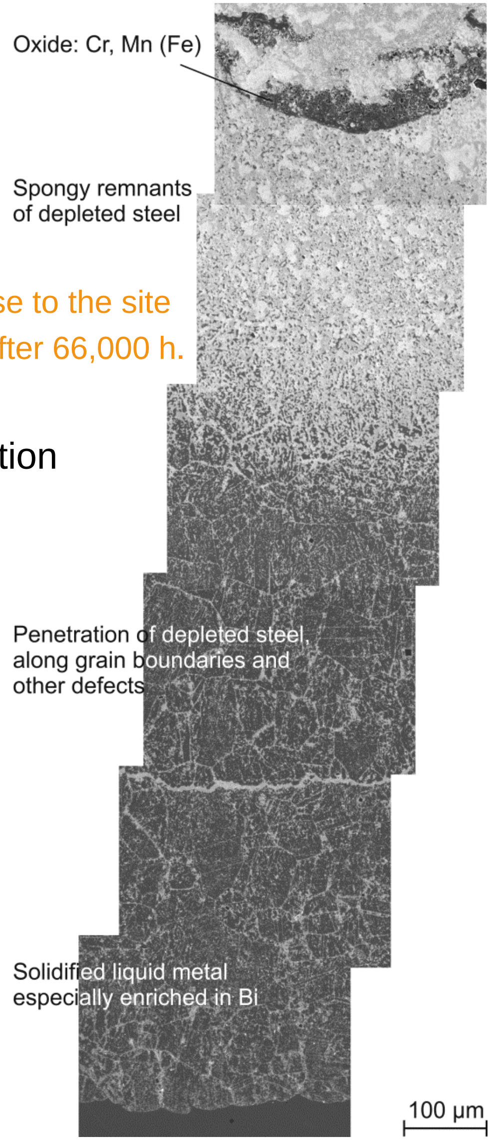
Sample T4 (left) and T10 (right) taken after operation for 40,000 and 108,000 h, respectively.



Mass flow during stage of partial plugging.

- Phenomena and associated degradation strongly depend on position.
  - Ranging from beneficial oxide scale formation to solution-based corrosion.
  - Preferential nickel (chromium) transfer to the LBE in cases of solution.

Remarkable local nickel release in Test-section 1 correlates in time with partial plugging of the loop!



Sample T6, close to the site of tube failure after 66,000 h.

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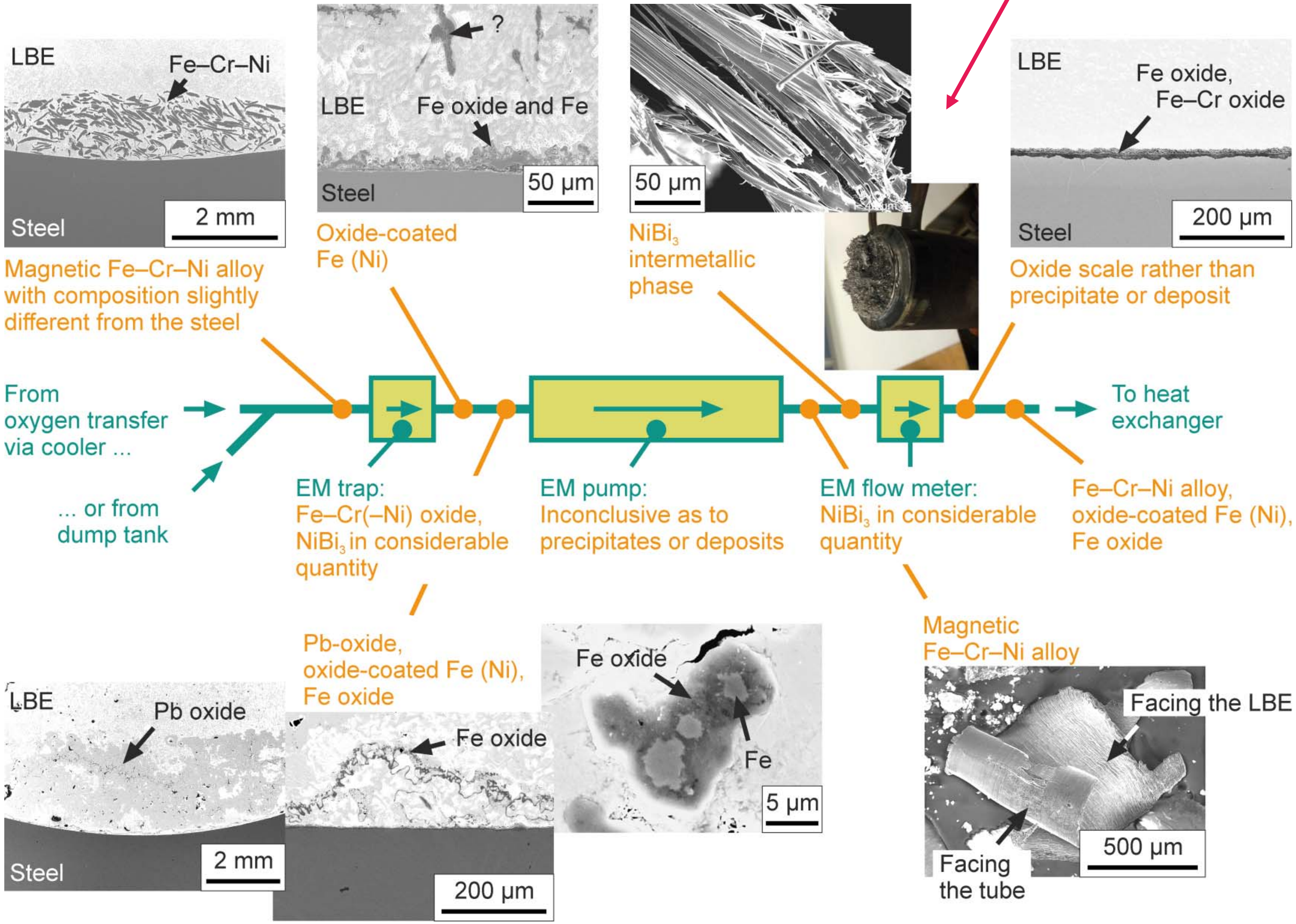
Precipitates or deposits in the cold leg

as found after operation for 113,000 h and attempts to refill the loop

The plug that prohibited refilling the loop after last LBE dump?



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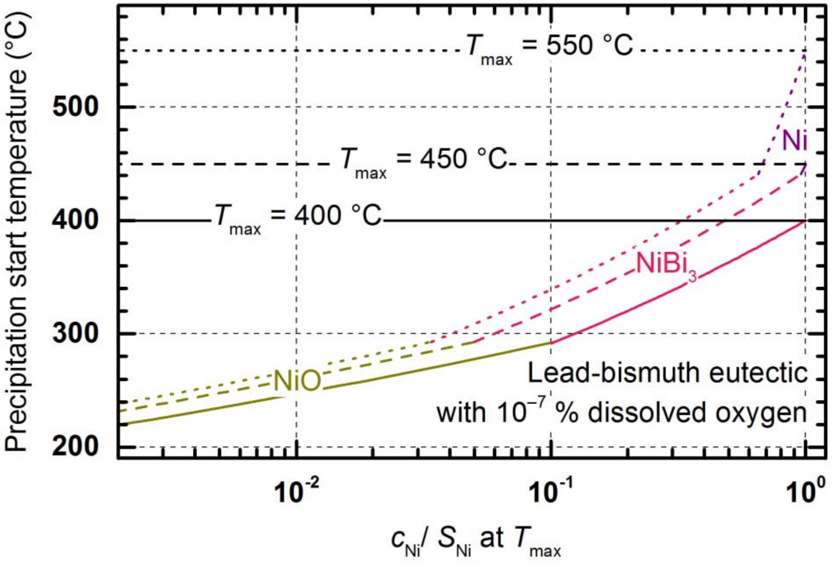
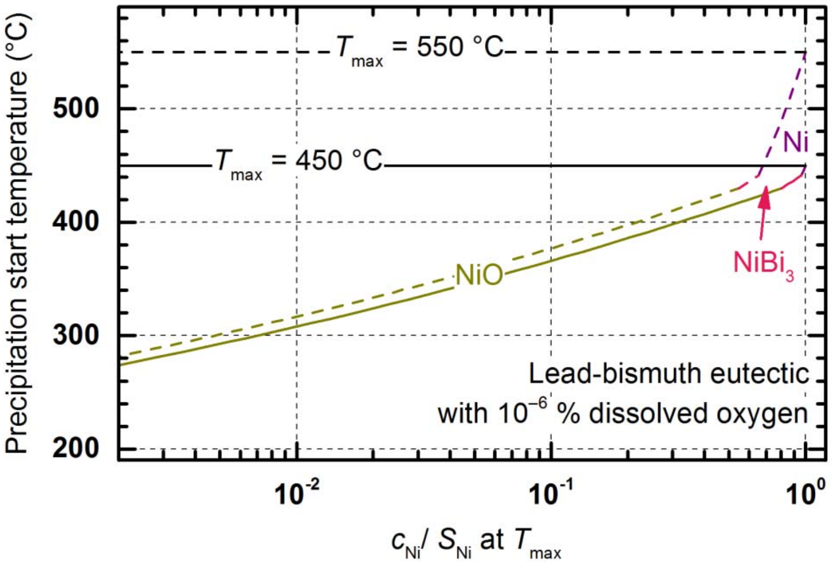
Nickel chemistry in the loop

- Simplified thermochemical calculations
  - Pure solid nickel.
  - Based upon experimental solubility S_{Ni} in the nickel and $NiBi_3$ domain.
  - Degree of nickel saturation of the LBE expressed as c_{Ni}/S_{Ni} at the maximum temperature along the loop.
  - Precipitation start temperatures for nickel, $NiBi_3$ or nickel oxide at 10^{-6} and 10^{-7} % oxygen dissolved in the LBE.



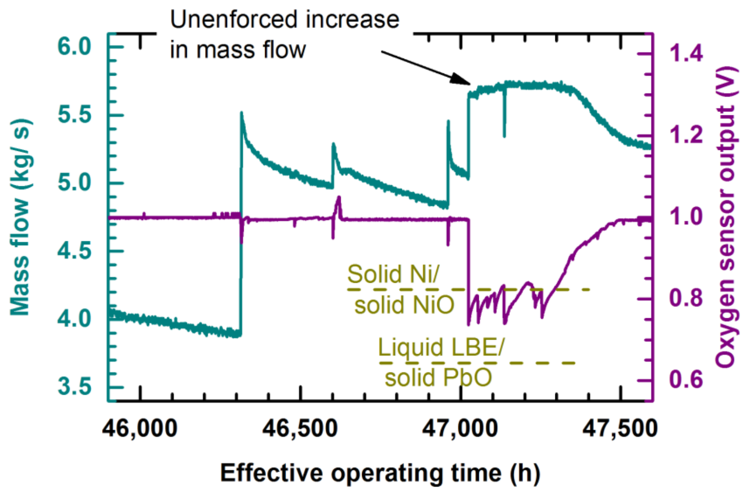


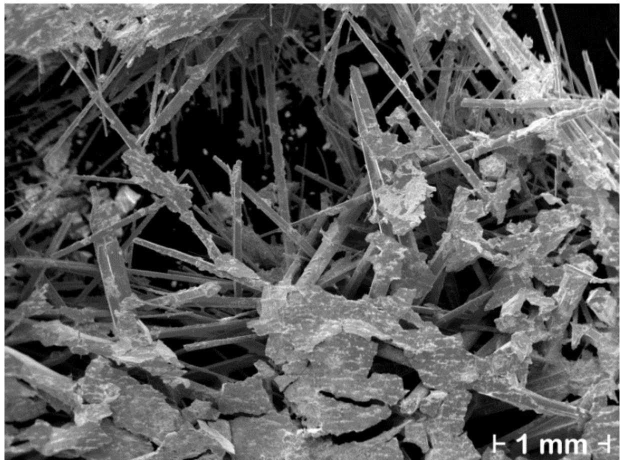
- Estimations for Grade 316 austenitic steel
  - c_{Ni}/S_{Ni} <1.
  - At 10^{-6} % oxygen, re-precipitation of nickel probably in the form of NiO.
  - At 10^{-7} % oxygen, $NiBi_3$ precipitation more likely. NiO only at temperatures below the minimum along the loop (~300–385 °C).

Observed plugging events

- Partial plugging started after 49,000 h of total operation
  - Accumulation of NiO in consequence of long-term operation at 10^{-6} % oxygen.
  - Eventual oxygen release inside the loop corroborates presence oxide (NiO or PbO).
  - Purging with argon–hydrogen gas improved the situation along with dumping the LBE and re-filling the loop.

Mass flow and dissolved oxygen during final stage of partial plugging.





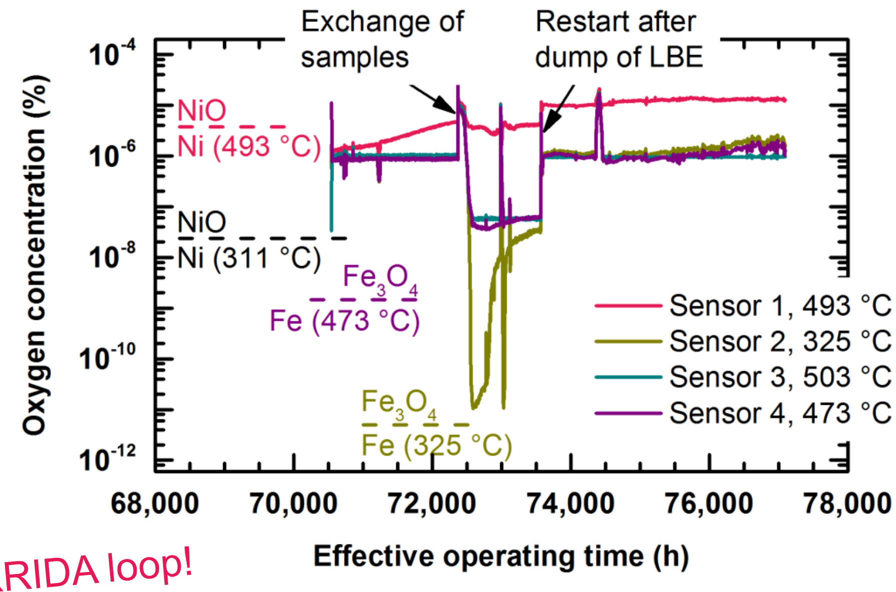
$NiBi_3$ collected from sensor port.

- $NiBi_3$ plug in the cold leg after 113,000 h, in connection with dumping of the LBE
  - Calculations suggest $NiBi_3$ formation during operation at 10^{-7} % oxygen.
  - $NiBi_3$ precipitation in cold spots like the EM trap, flow meter, sensor and specimen ports.
  - Oxidation of $NiBi_3$ possibly aggravates the mass flow issue after the return to 10^{-6} % oxygen.
  - Plug formed from $NiBi_3$ in the flow meter that was mobilised by dumping the LBE.

Interference of dissolved steel elements …

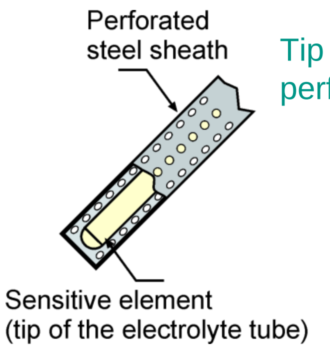
- … with oxygen control
  - Oxidation of nickel (dissolved or $NiBi_3$) in the cold leg.
  - Dissolved oxygen concentration after transfer rises in order to compensate for the consumption.
  - This concentration is limited by NiO formation at the temperature of oxygen transfer.
  - Eventually, NiO formation in the cold leg determines the concentration of dissolved oxygen in the hot leg.

Indicated concentration of dissolved oxygen for experiments at nominally 500 °C/ 10^{-6} % oxygen.



Specific to the CORRIDA loop!

- … with oxygen measurement
  - Especially the sensor in the cold leg occasionally indicates by orders of magnitude lower oxygen concentration.
  - Re-precipitated iron (or elements forming more stable oxides than iron) collects at the sensitive element and oxidizes.



Tip of oxygen sensor with perforated steel sheath.

Estimation of the long-term reliability of oxygen sensors

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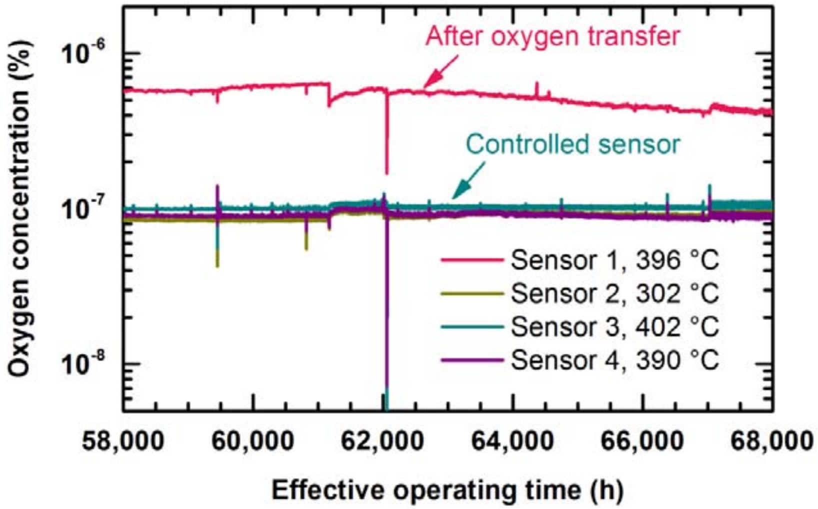
- Acceptance test before installation to the loop
  - Qualitative tests in the past.
  - Quantitative tests in the future.

- Repeated test after some time of operation hardly possible
  - Difficult if not impossible to adjust an appropriate metal/ metal oxide equilibrium in the loop.
  - High risk that sensor breaks when being removed from the loop.

- **Criterion for proper long-term operation in the CORRIDA loop**
  - Compare the concentration of oxygen calculated from the four sensor signals.
  - Take into account oxygen consumption, accuracy of converting indicated voltage into oxygen concentration or temporary effects of sensor fouling.



Comparison of sensor output during steady operation at 400 °C/ 10^{-7} % dissolved oxygen.

Conclusions

- LBE loops primarily made of austenitic steel:
  - In the presence of oxygen, plugging is dominated by dissolved nickel.
  - The deposited nickel compound changes between 10^{-7} and 10^{-6} % dissolved oxygen from $NiBi_3$ to NiO.
  - Needle-shaped $NiBi_3$ crystals of several mm length tend to plug the tubing when mobilized, e.g., by dumping the LBE.
  - NiO seems the more preferable deposit in comparison to $NiBi_3$.

- Oxygen transfer to flowing LBE:
  - In the long run, oxygen consumption and release in the loop are equally important.
  - Noticeable interference of dissolved nickel possible, especially in the case of the CORRIDA loop.
  - Gas/ liquid transfer generally meets the variable request for oxygen addition or removal.

- Electrochemical oxygen sensors:
  - Sensors with Pt/ air reference electrode operate reliably in the loop for >90,000 h at temperatures between ~400 and 550 °C.
  - Fouling of sensors, with temporary effect on the measurement, attributable to precipitated iron (or other elements forming more stable oxides than iron).

Acknowledgements

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Thank you for your attention!