Challenges of materials qualification for nuclear systems with heavy liquid metal coolant:

Effect of LBE on materials properties

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MYRRHA

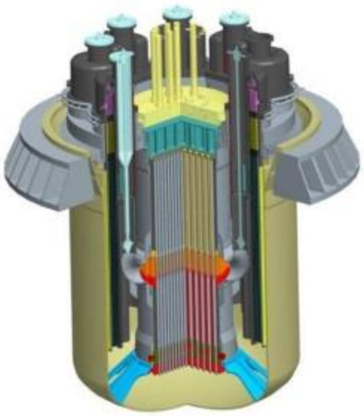
International workshop "Materials resistant to extreme conditions for future energy systems", June 12-14, 2017, Київ, Україна

Overview

- Heavy liquid metal coolants
- Role of material research for development of MYRRHA
- Materials degradation effects
  - Liquid Metal Corrosion (LMC)
  - Liquid Metal Embrittlement (LME)
  - Irradiation effects
  - Synergetic effect
- Summary

Heavy liquid metal coolant

- Neutronics, related to the fast spectrum necessary for breeding, fuel conversion and actinide transmutation
- No violent exothermic reaction with water/air
- A very high boiling temperature, reducing the risk for loss of coolant
- An excellent potential for decay heat removal by natural convection
- Inherent shielding of gamma radiation from fission products
- Target materials for high-power neutron spallation sources (ADS)



ALFRED

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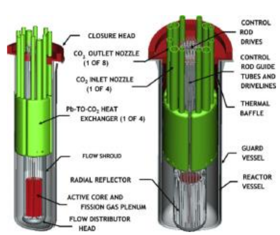
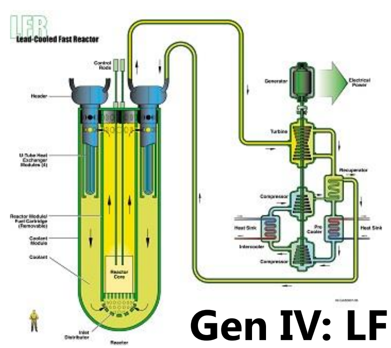
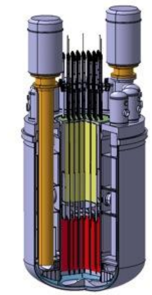


SMR

SSTAR



Gen IV: LFR



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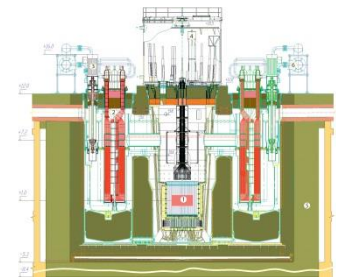
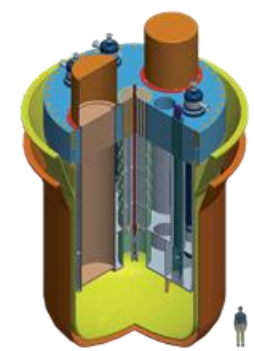


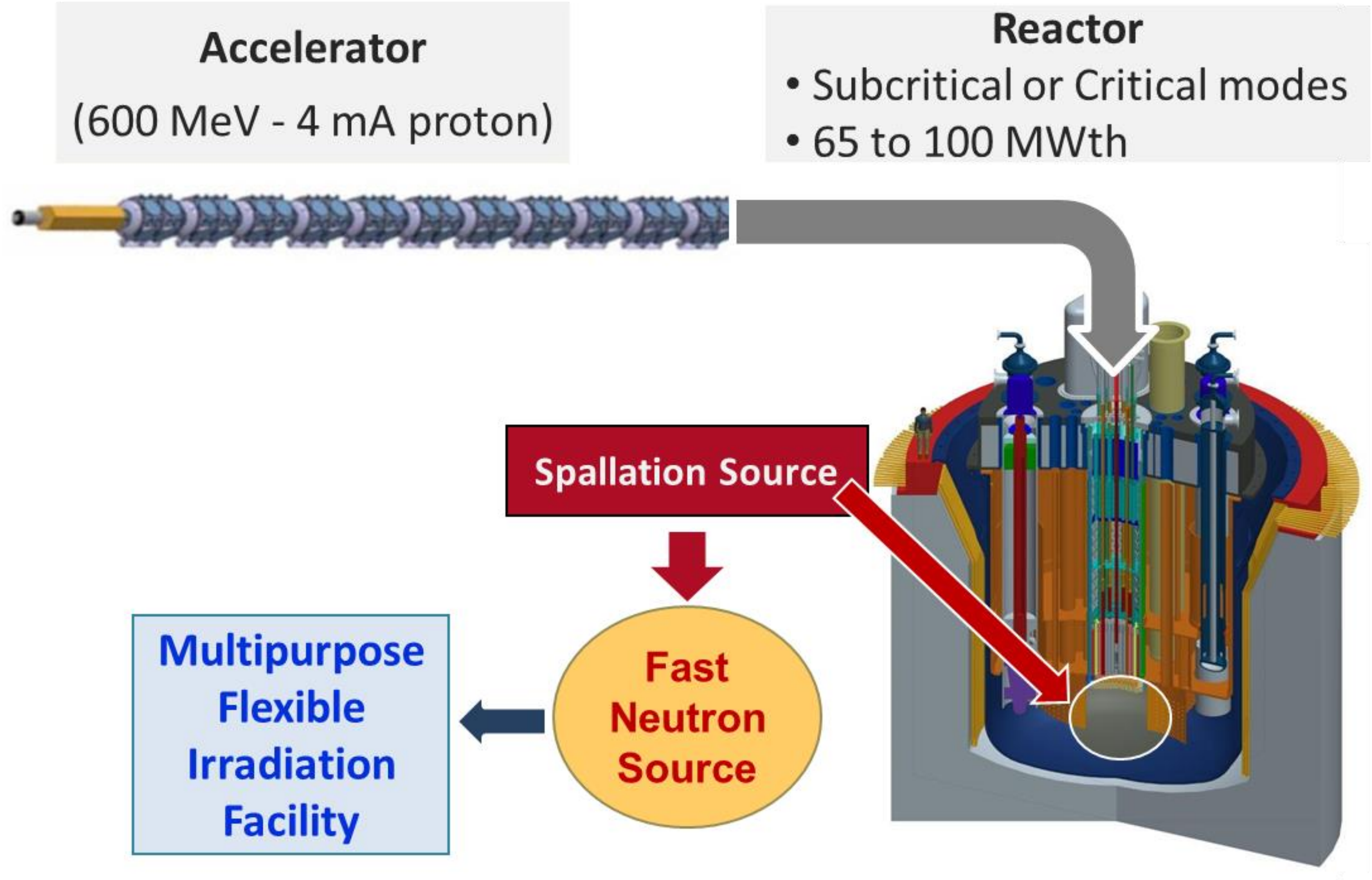
BREST-OD-300



MYRRHA

MYRRHA

- MYRRHA: accelerator driven system (ADS)

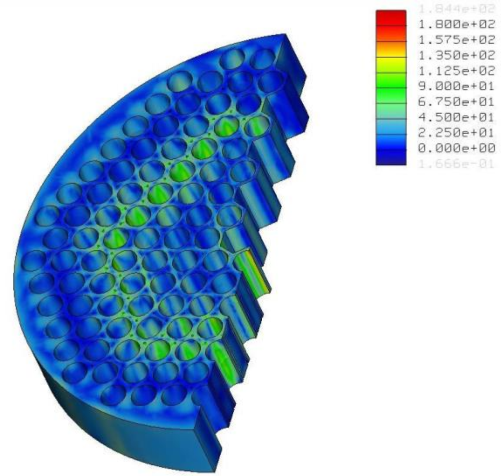


Role of material research for development of MYRRHA

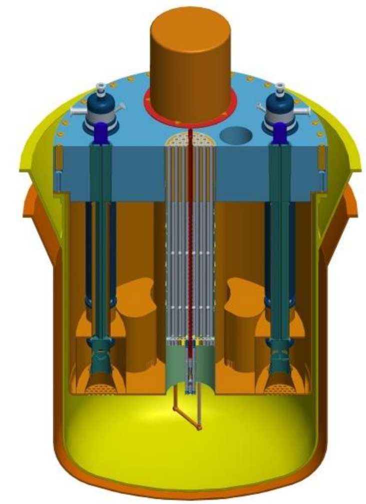
Design tools:

- Nuclear manufacturing code: RCC-MRx
- Fuel codes
- FE calculations

Design

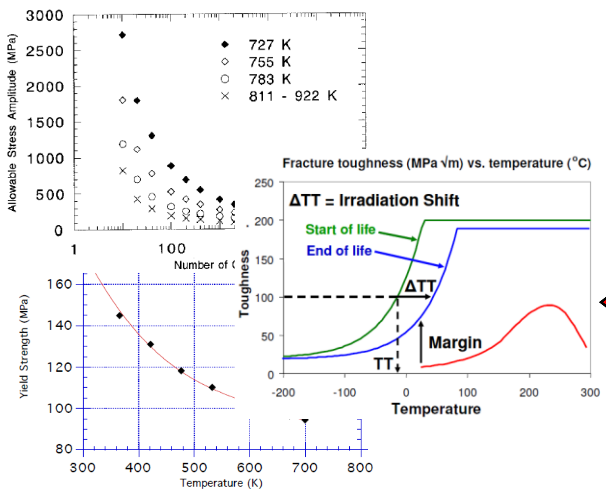


Construction



Approaches

Materials properties



Data

Required data for MYRRHA:

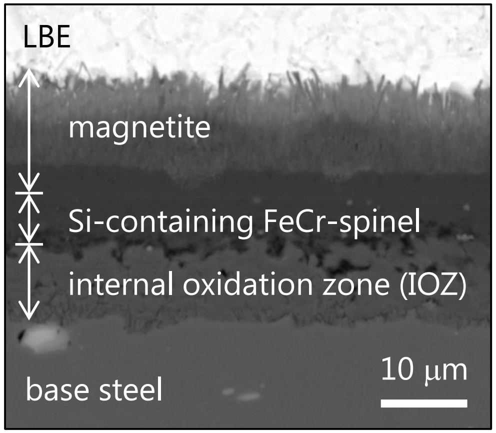
- Some basic characteristics of candidate materials (T91, SS 316L & 1.4970)
- Effects of LBE & irradiation on material properties
- Physical effects: Liquid metal embrittlement (LME), Liquid Metal Corrosion (LMC), SCC, etc.

Materials degradation effects to be investigated

- Liquid Metal Corrosion (LMC)
- Liquid Metal Embrittlement (LME)
- Irradiation effects
- Synergetic effects

Liquid Metal Corrosion

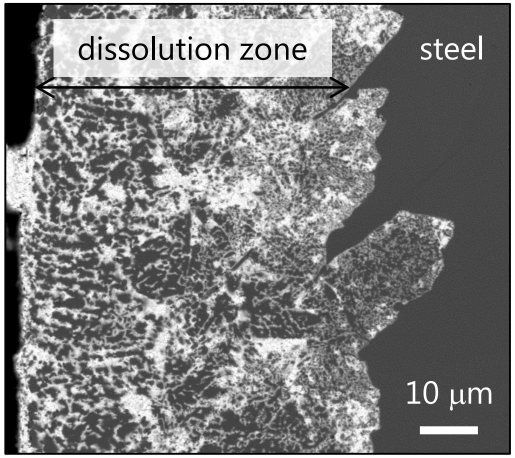
1. Oxidation



- Multi-layered oxide scales form in contact with O-containing LBE on steel surface
- If protective at service conditions, oxide scales minimize further attack of steel by LBE

EP-823: 490°C, 5016 h, oxygen saturation, static LBE
(K. Lambrinou, SCK•CEN data)

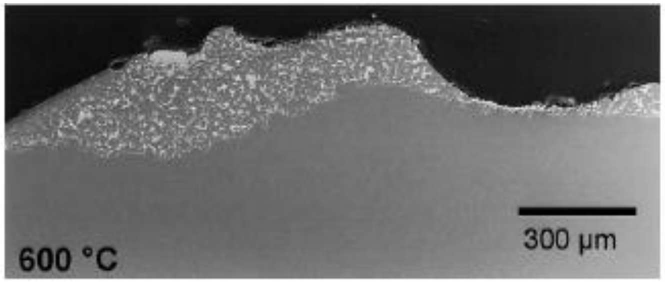
2. Dissolution



- Loss of steel alloying elements (Ni, Mn, Cr)
- LBE penetration
- Ferritization of dissolution zone due to loss of austenite stabilizers (Ni, Mn)

316L: 500°C, 3282 h, $7.5\times10^{-13} < C_O$ (wt%) $< 2.8\times10^{-8}$, static LBE *(K. Lambrinou, SCK•CEN data)*

3. Erosion



- Severe material loss & compromise of structural integrity
- Observed at high LBE flow velocities, two-phase flow, and sites of flow diversion

316L: 600°C, 2000 h, $C_O \approx 10^{-6}$ wt%, flowing LBE ($v \approx 2$ m/s)
*(Müller et al., Journal Nuclear Materials, **301** (2002) 40-46)*

Effects of Corrosion on Reactor Operation

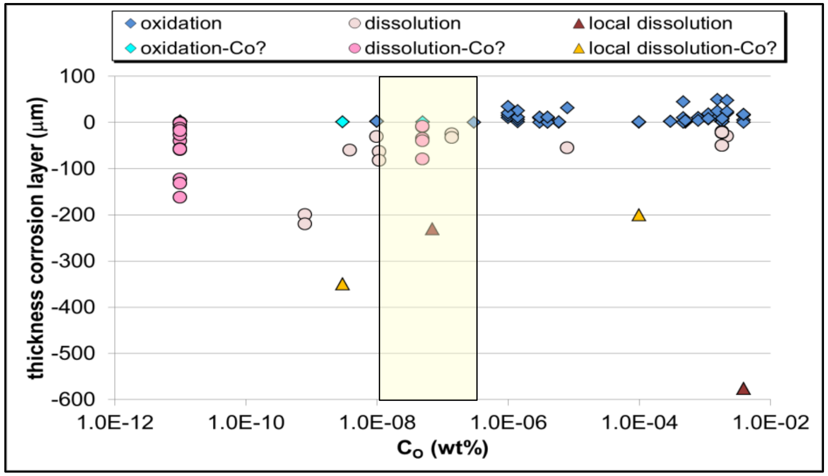
Possible Effects of Corrosion on Reactor Operations

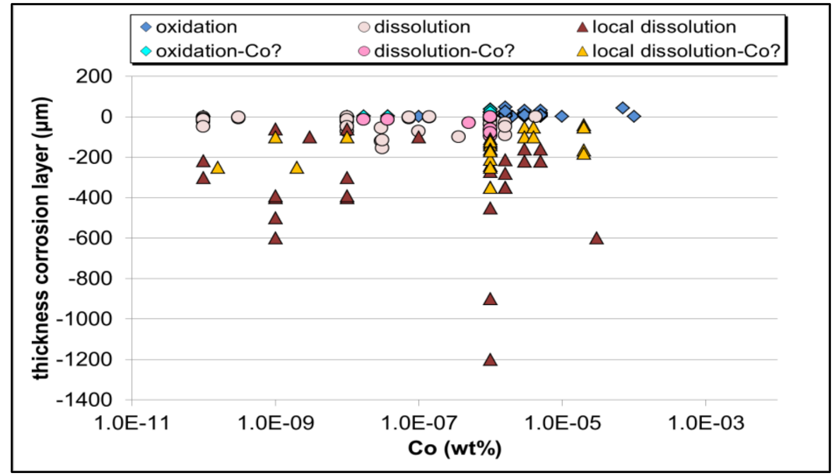
- Material loss (dissolution, erosion) → component integrity
- Change in thermal conductivity (oxidation) → change of heat transfer characteristics
- Plugging due to deposition of corrosion products → flow obstruction

Principal directions of corrosion program

- Development of corrosion correlations for design (deterministic ↔ empiric approach)
  - Boundary operating conditions and a little bit beyond
    - For oxidation ([O]↑, T↑, v↓)
    - For dissolution & erosion ([O]↓, T↑, v↑)
- Investigation of oxide layer properties
  - Maximum and average thicknesses
  - Thermal conductivity
- Assessment of corrosion products release to the coolant and oxygen consumption

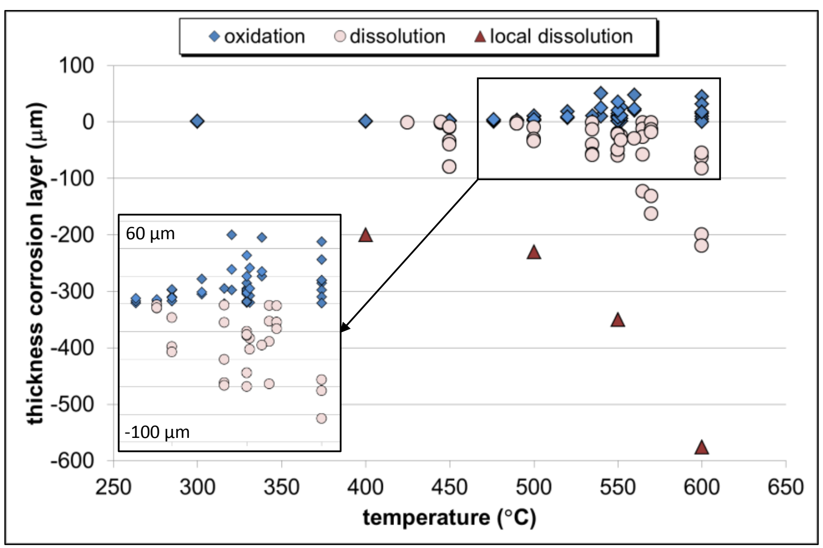
Literature data on corrosion of 316L in LBE

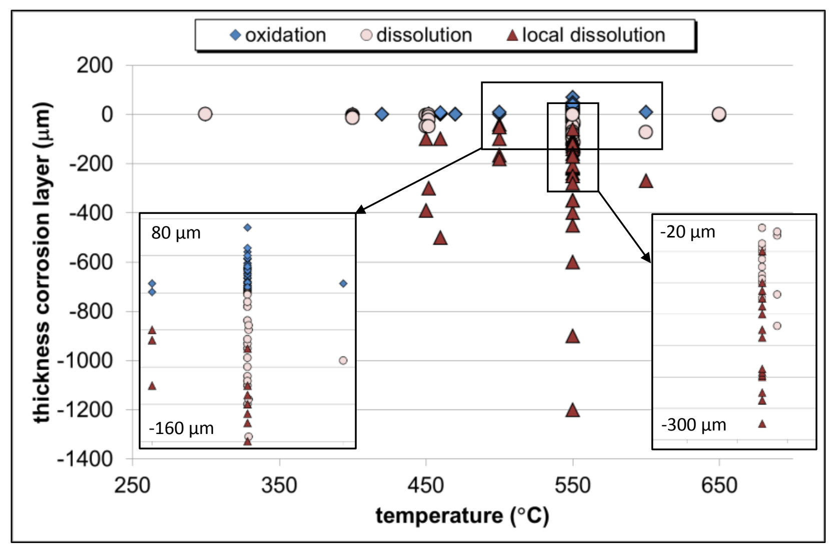




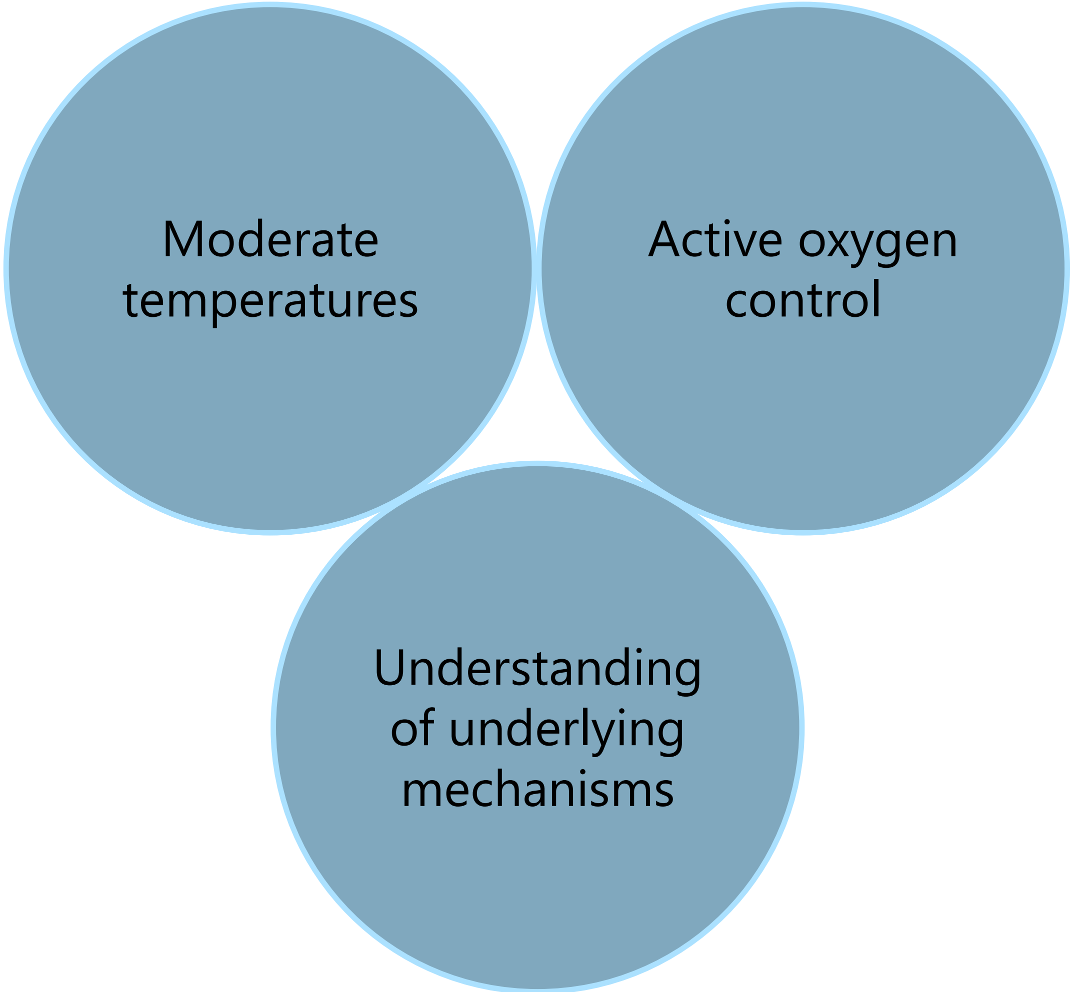
Static / quasistatic LBE

Flowing LBE





Three pillars of Liquid Metal Corrosion mitigation strategy



Solutions for LMC

- "Short term" solution
  - Investigation of LMC in depth **FP6 EUROTRANS FP7 GETMAT FP7 MATTER**
  - Database on corrosion for candidate materials including welding joints **H2020 GEMMA**
  - Design correlations to incorporate LMC
  - Adjustments of the reactor parameters
    - Temperature range
    - Components lifetime
    - Development of inspection and surveillance programs
- Midterm solution
  - Surface alloying to create protective barrier **FP6 EUROTRANS FP7 GETMAT**
  - Weld overlay
  - Coatings **FP7 GETMAT**
  - Qualification for application on functional & structural components **H2020 GEMMA**
- Long term solution
  - Development of nuclear grade corrosion resistant materials **H2020 GEMMA**
    - FeCrAl **H2020 GEMMA**
    - Alumina Forming Austenitic Steels (AFA) **H2020 GEMMA**

Materials degradation effects to be investigated

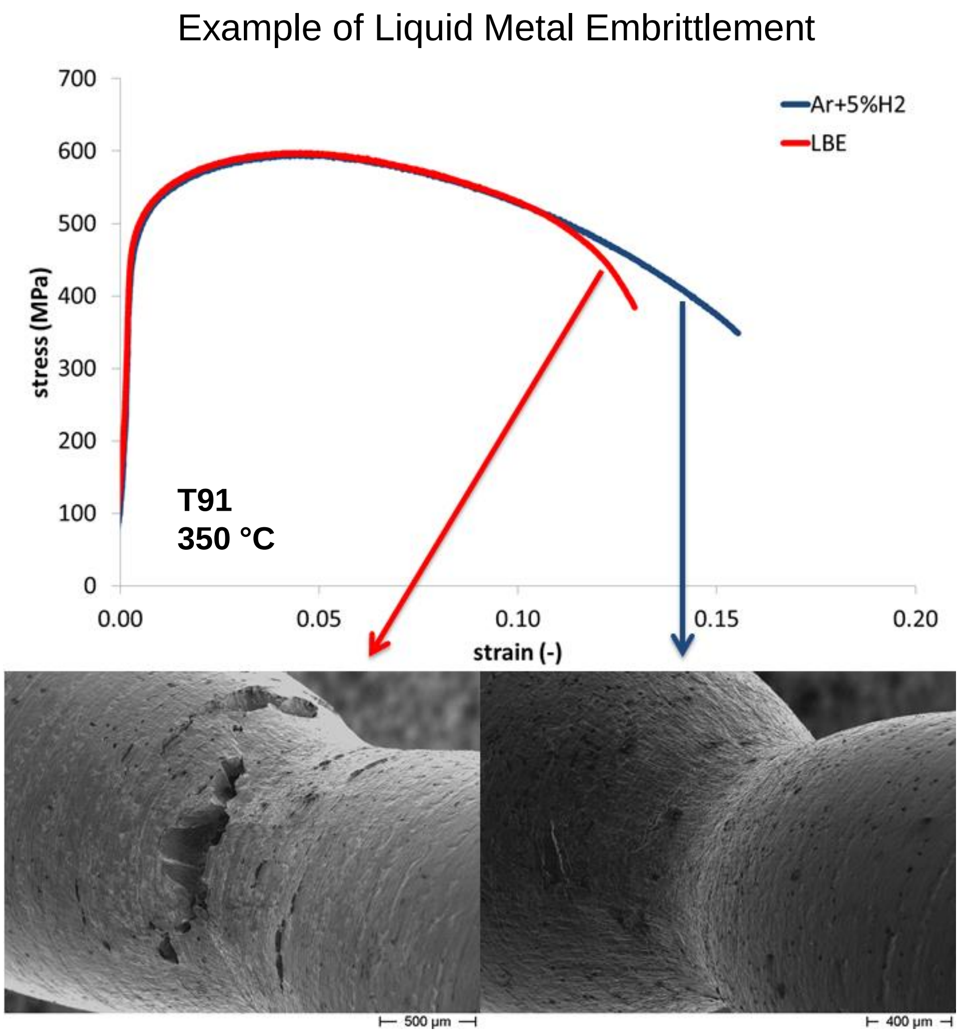
- Liquid Metal Corrosion (LMC)
- Liquid Metal Embrittlement (LME)
- Irradiation effects
- Synergetic effects

Liquid Metal Embrittlement (LME) effect

Degradation of steel's mechanical properties in contact with liquid metal

Potentially can affect:

- Tensile properties
  - Total elongation
- Fracture toughness
- Fatigue properties
  - Endurance
  - Crack Growth Rate
- Creep properties
  - Creep rate
- Creep-fatigue properties



The earliest documented observation of Liquid Metal Embrittlement

"A piece of galvanized iron wire, of good quality, such that when cold it could be bent several times on itself and back again before breaking, was raised to a red heat so quickly that the coating of zinc was melted and only a small portion vaporized. On attempting to bend it whilst still red-hot, it broke off sharp, offering very little resistance to fracture. The fracture was of a uniform blue-grey colour, as though the zinc had penetrated into the interior of the iron. When cold, the same piece broke with all its former toughness and with a long fibrous fracture. The wire was again heated till the coating of zinc was completely vaporized, and then it was found to be so tough that it was impossible to break at a red heat. Wire in red-hot molten zinc will often break short, though the part out of the metal remains quite tough."

Johnson W.H., On Some Remarkable Changes Produced in Iron and Steel by the Action of Hydrogen and Acids, Proceedings of the Royal Society of London (1854-1905), **1874**. 23: p. 168-179

Investigations of LME in Ukraine

УДК 532.6:539.4

І. Г. ДМУХОВСЬКА, В. В. ПОПОВИЧ

ВПЛИВ КОНЦЕНТРАТОРІВ НАПРУЖЕНЬ НА ТЕМПЕРАТУРНУ ЗАЛЕЖНІСТЬ РІДКОМЕТАЛЕВОГО ОКРИХЧЕННЯ АРМКО-ЗАЛІЗА

На даний час можна вважати встановленим, що рідкометалеве окрихчення (РМО) сталей адсорбційно діючими розплавами відбувається в обмеженому інтервалі температур [1—5], який тісно пов'язаний

Академия наук Украинской ССР

Физико-механический институт им. Г.В.Карпенко

В.В.ПОПОВИЧ, И.Г.ДМУХОВСКАЯ

ЖИДКОМЕТАЛЛИЧЕСКОЕ ОХРУПЧИВАНИЕ ДЕФОРМИРУЕМЫХ МЕТАЛЛОВ

Препринт № 69

ФИЗИКО-ХИМИЧЕСКАЯ МЕХАНИКА МАТЕРИАЛОВ 1968, том 4, № 1

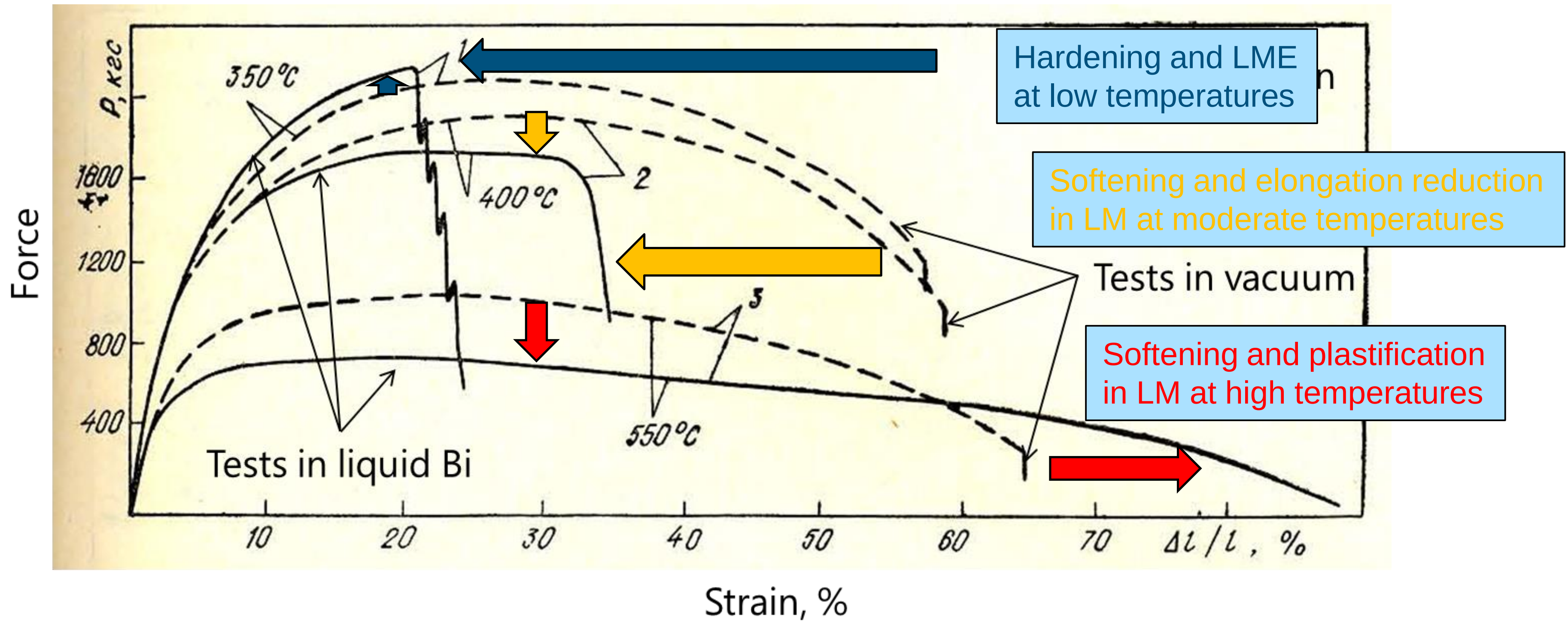
УДК 620.172:539.431

ВЛИЯНИЕ РАСПЛАВОВ ЛЕГКОПЛАВКИХ МЕТАЛЛОВ НА УСТАЛОСТНУЮ ПРОЧНОСТЬ УГЛЕРОДИСТОЙ СТАЛИ В ЗАВИСИМОСТИ ОТ ИЗМЕНЕНИЯ ЧАСТОТЫ НАПРЯЖЕНИЯ

Е. С. НИКОЛИН, Г. В. КАРПЕНКО

(Физико-механический институт АН УССР, Львов)

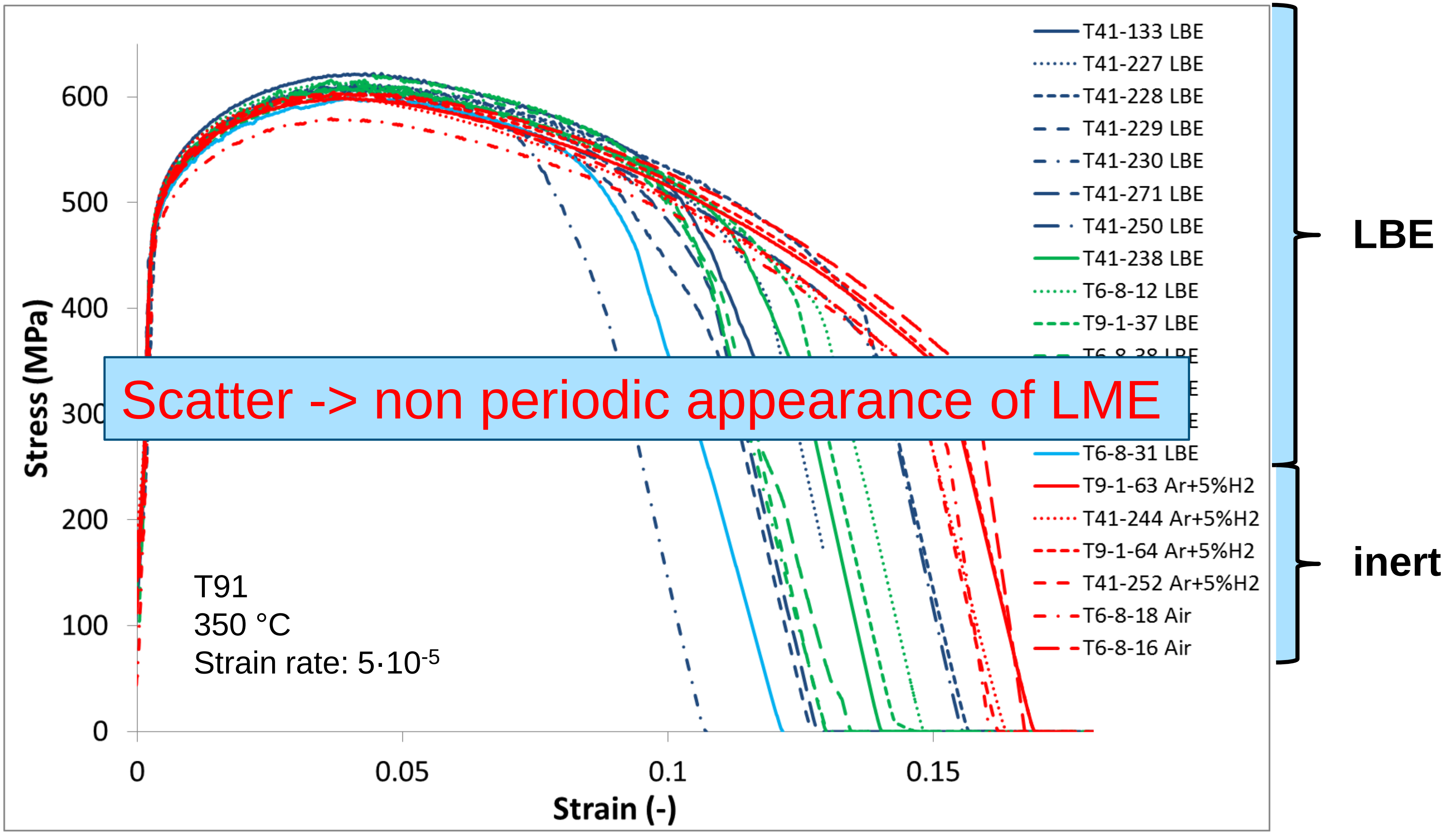
Armco iron in liquid bismuth



Stress-strain curves of Armco iron specimens tested in bismuth (continuous lines) and in vacuum (dashed lines) at 350 °C, 400 °C and 550°C

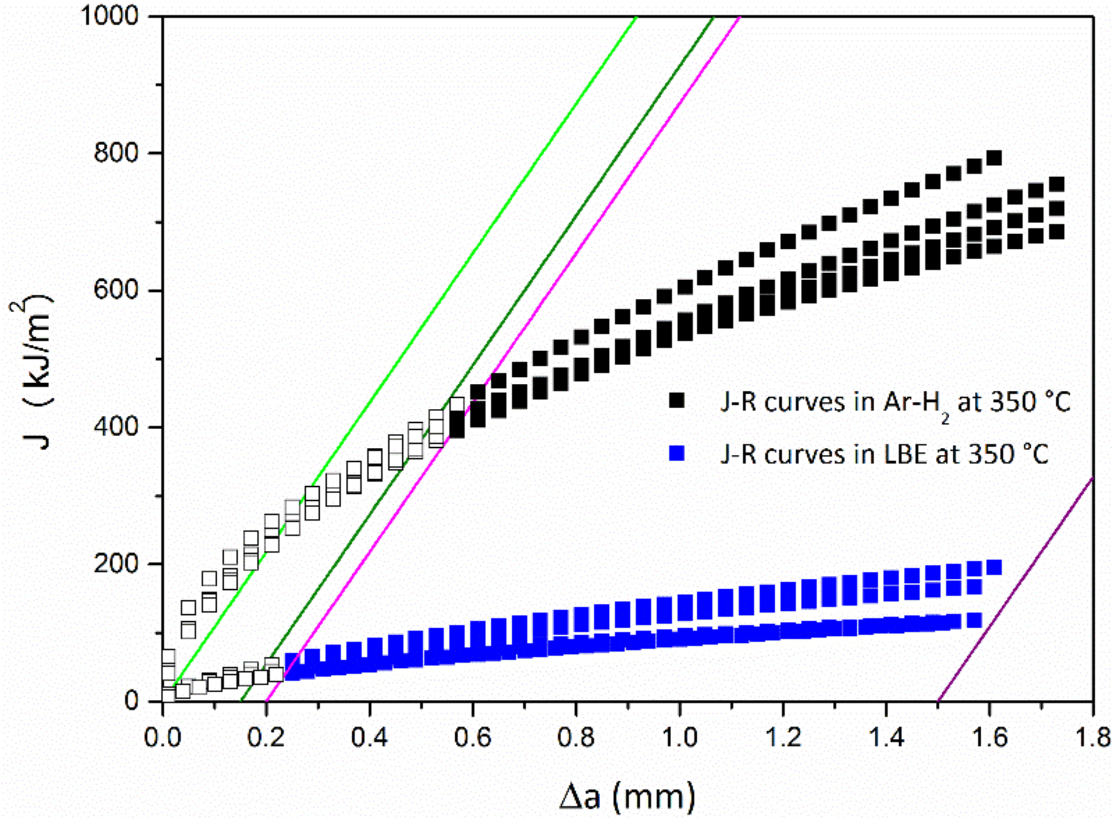
Popovich, V.V. and I.G. Dmukhovskaya, Rebinder effect in the fracture of Armco iron in liquid metals. Soviet materials science : a transl. of Fiziko-khimicheskaya mekhanika materialov / Academy of Sciences of the Ukrainian SSR, 1978. 14(4): p. 365-370.

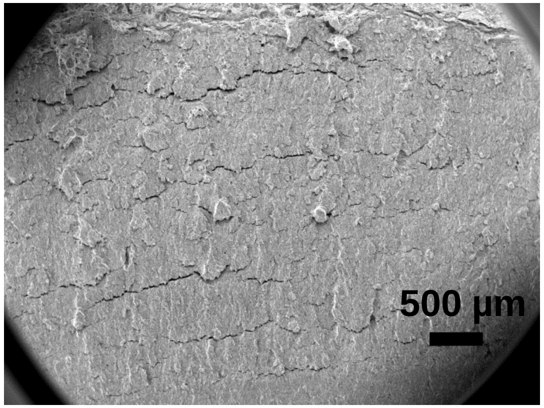
T91 in LBE: Tensile data

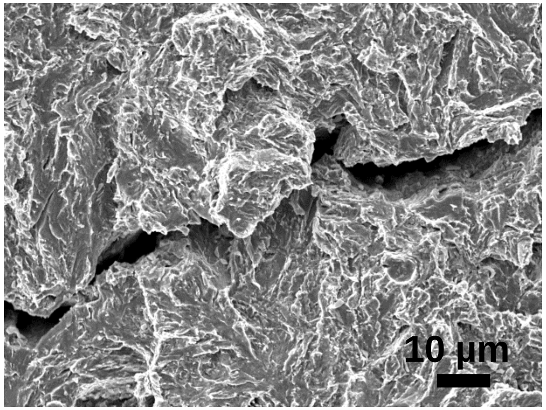


Stress-strain curves for slow strain rate tests, at $5 \cdot 10^{-5} s^{-1}$, of **T91 steel** in Ar+5%H_2 and in LBE containing $10^{-9} \div 10^{-10}$ wt.% of dissolved oxygen, at 350 °C.

Fracture toughness tests in LBE



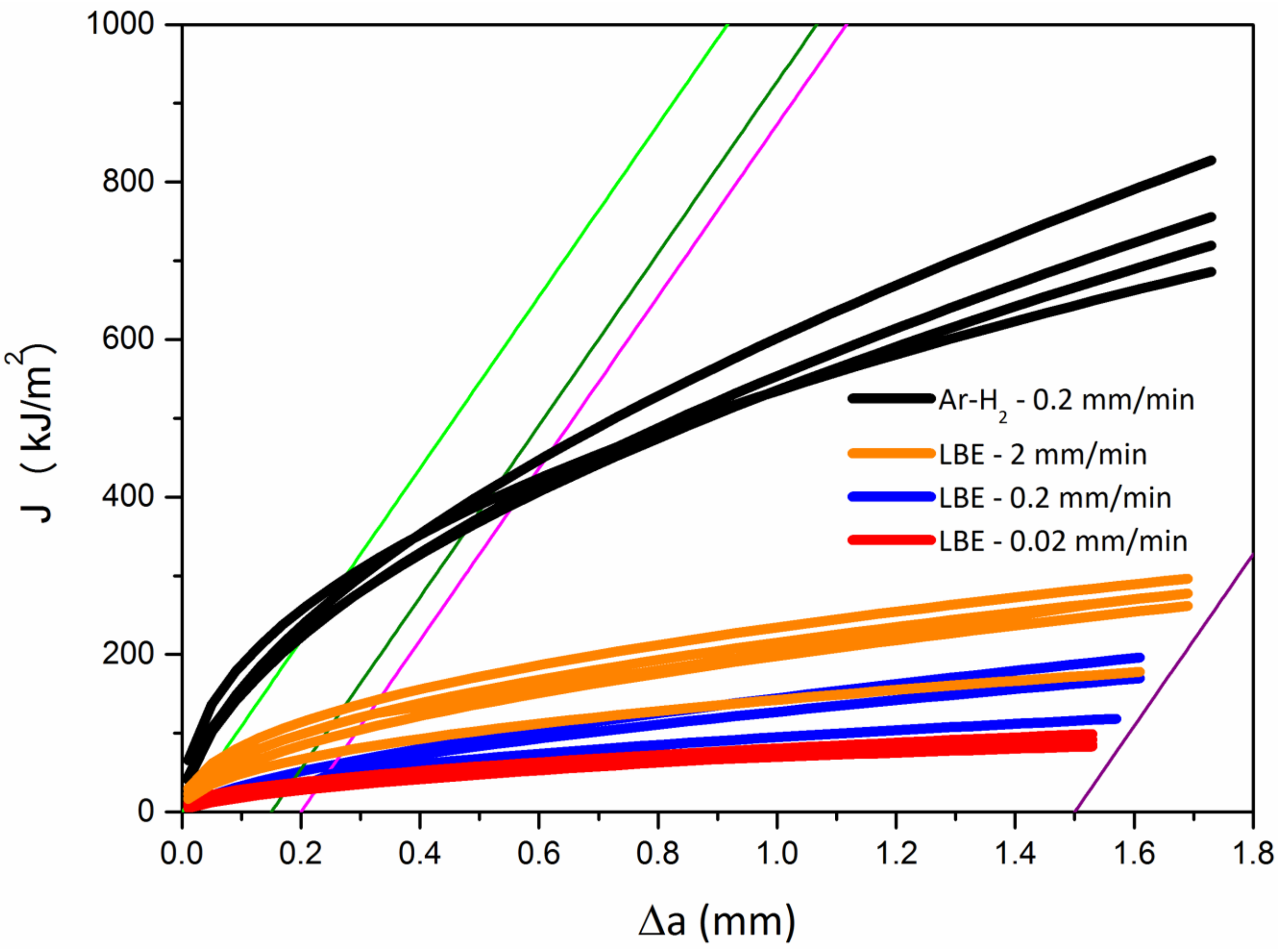


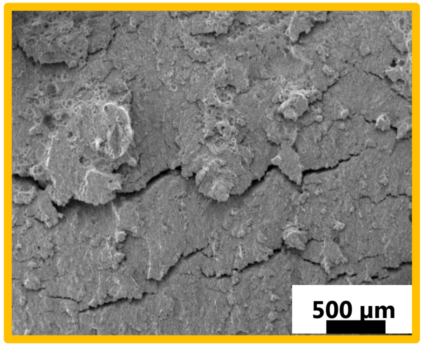


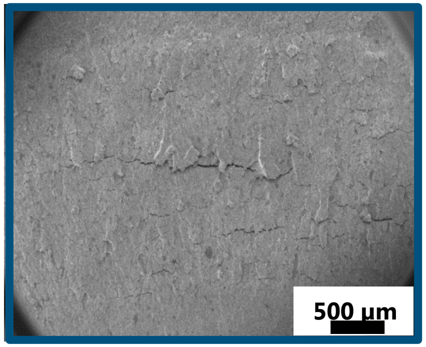
J-a curves of **T91 steel** specimens in air and in LBE containing $10^{-9} \div 10^{-10}$ wt.% of dissolve d oxygen, at 350 °C.

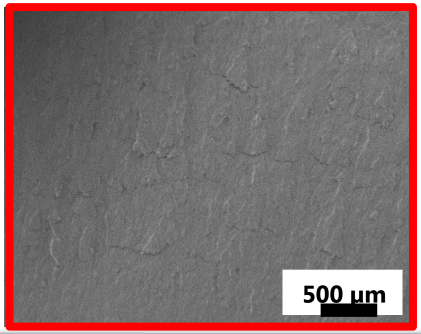
Feyzan Ersoy, Serguei Gavrilov, Kim Verbeken, Investigating liquid-metal embrittlement of T91 steel by fracture toughness tests, Journal of Nuclear Materials, Volume 472, 2016,171-177

Effect of displacement rate on FT

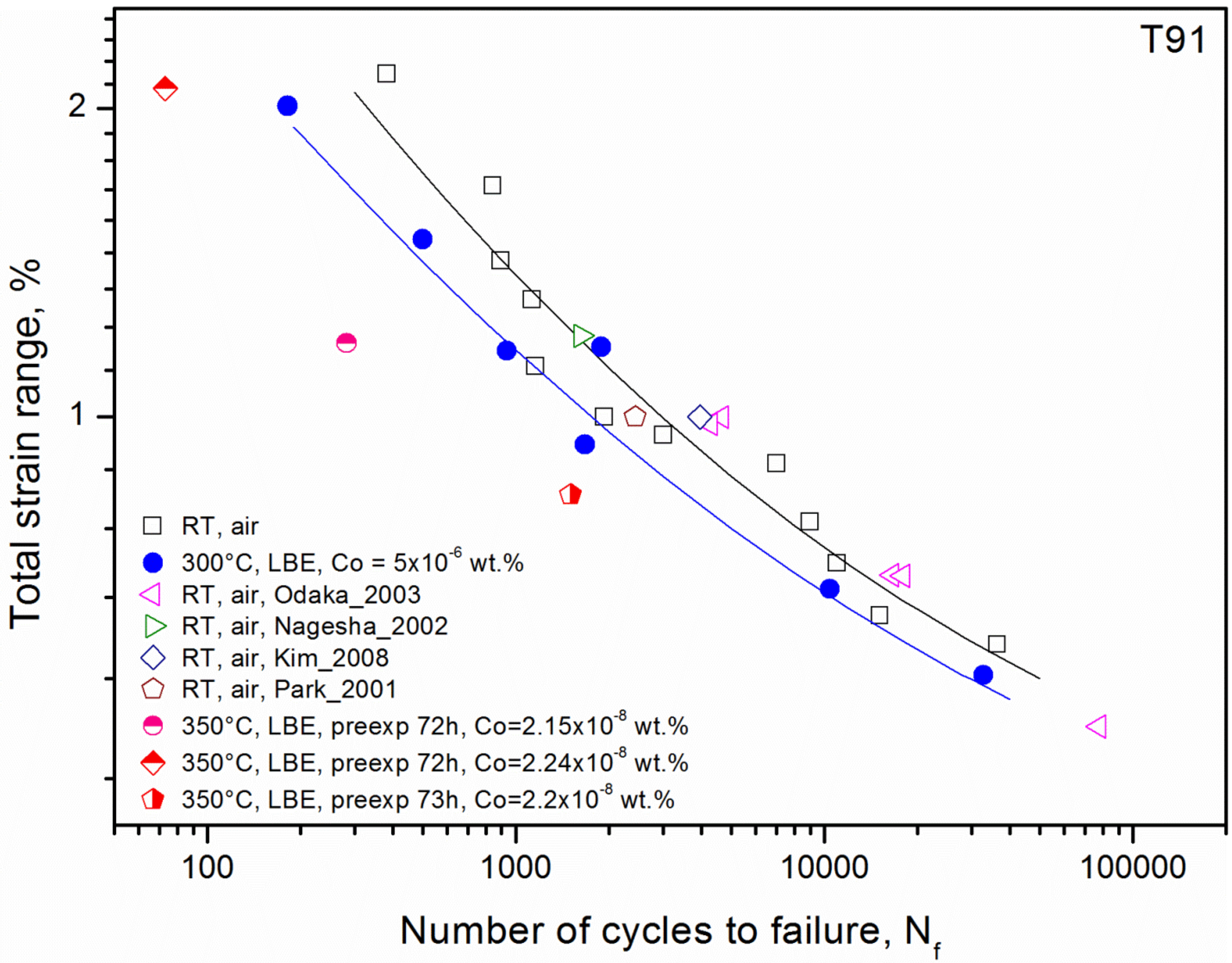








Fatigue endurance

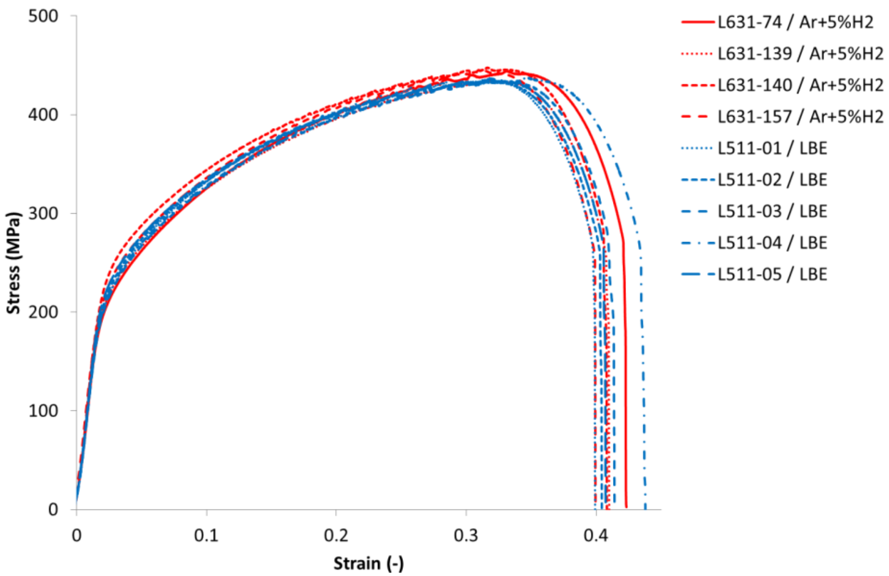


Fatigue endurance diagram of **T91 steel** in air and in LBE at RT and 350 °C.

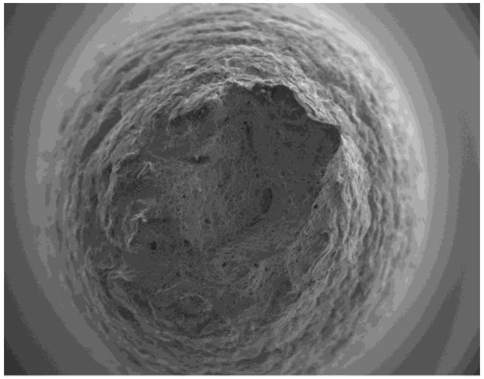
X.Gong,P.Marmy,L.Qin,B.Verlinden,M.Wevers,M.Seefeldt, Effect of liquid metal embrittlement on low cycle fatigue properties and fatigue crack propagation behavior of a modified 9Cr-1Mo ferritic-martensitic steel in an oxygen-controlled lead-bismuth eutectic environment at 350 °C, Materials Science&Engineering A 618 (2014) 406-415

X.Gong,P.Marmy,B.Verlinden,M.Wevers,M.Seefeldt, Low cycle fatigue behavior of a modified 9Cr-1Mo ferritic-martensitic steel in lead-bismuth eutectic at 350°C – Effects of oxygen concentration in the liquid metal and strain rate, Corrosion Science 94 (2015) 377-391

Investigation of susceptibility of 316L

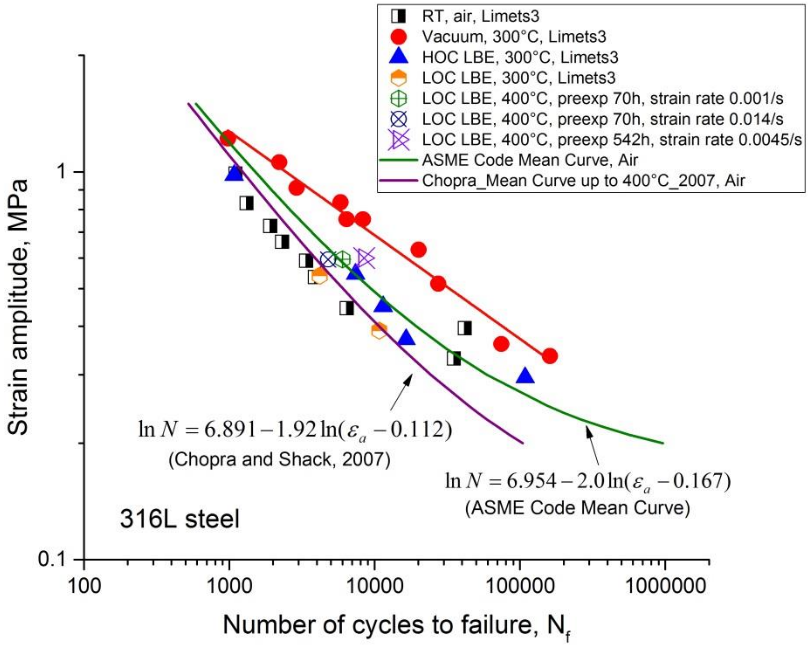


SSRT



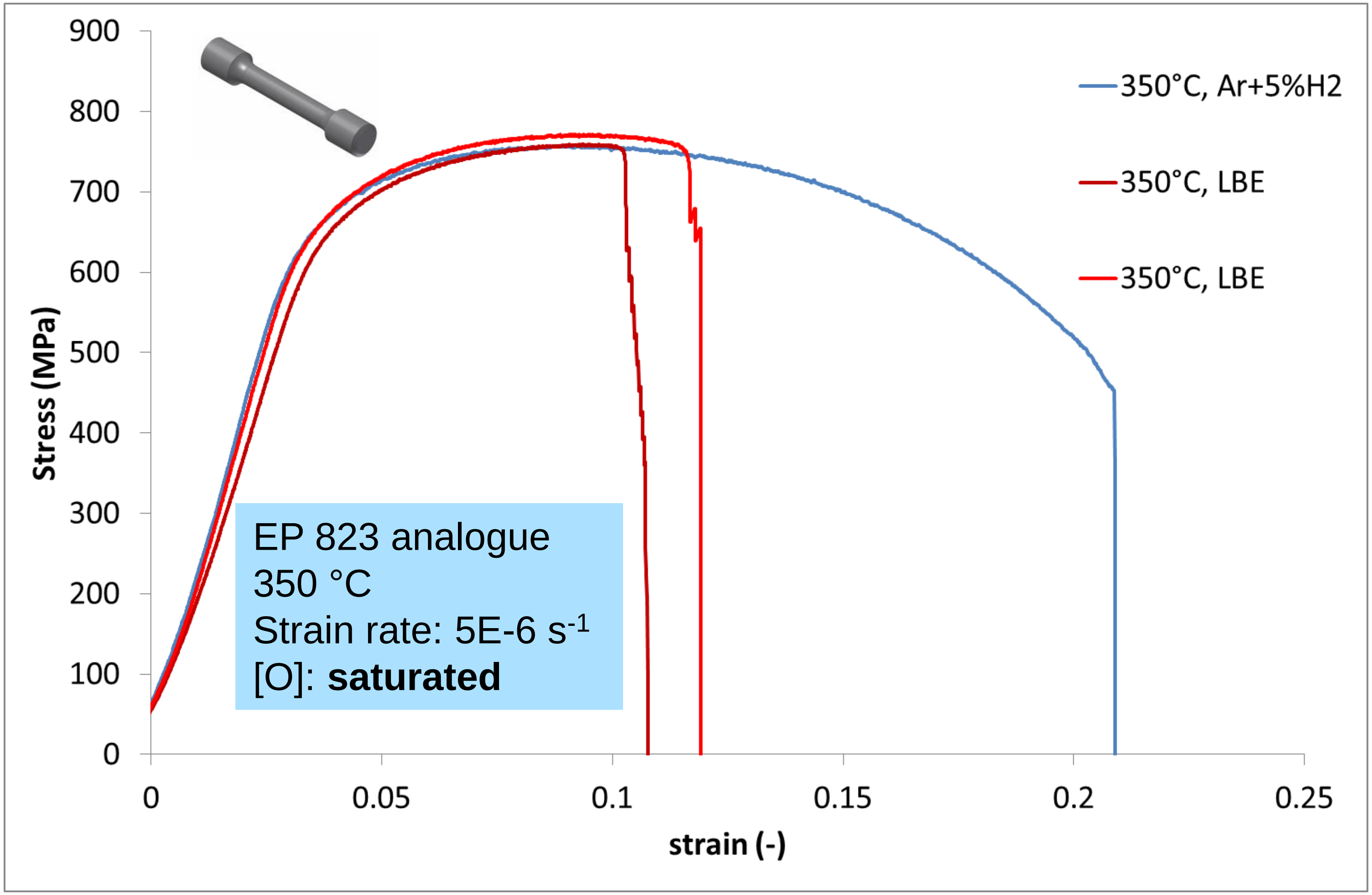
⊢ 600 µm ⊣

Fractography



Fatigue

LME of EP823 analogue

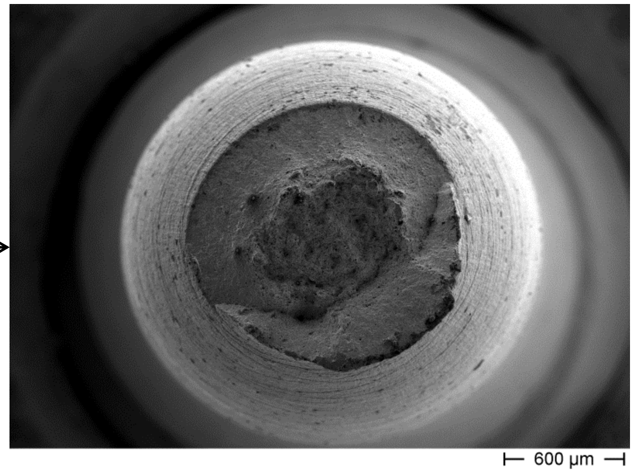


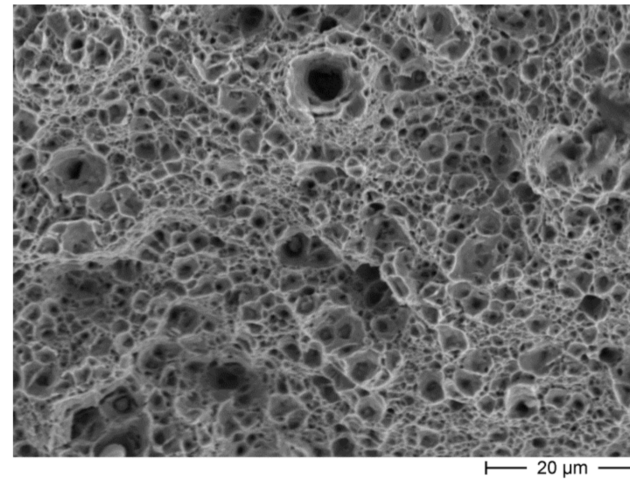
Van den Bosch, J., et al., *On the LME susceptibility of Si enriched steels.* Journal of Nuclear Materials, 2012. **429**(1–3): p. 105-112.

EP823 analogue: Fractured surface

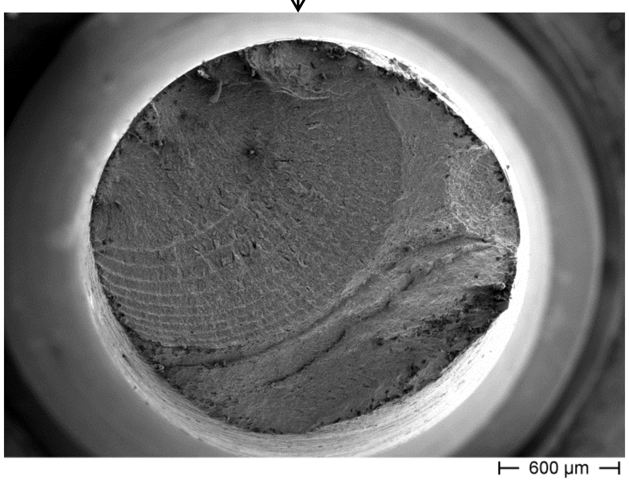
EP823 analogue
350 °C
Strain rate: 5E-6 s^{-1}

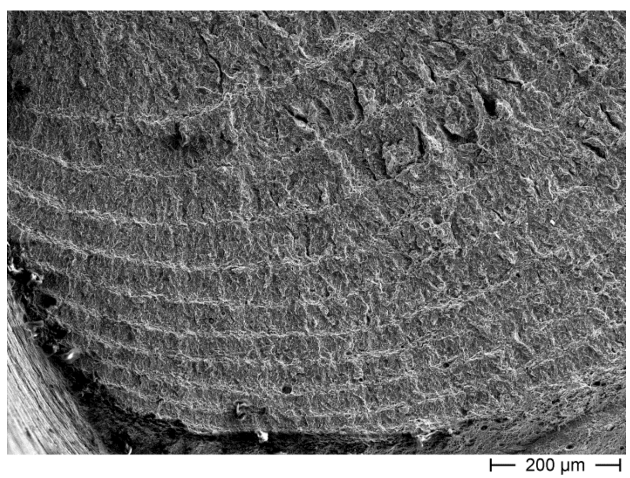
Ar+5%H2 →

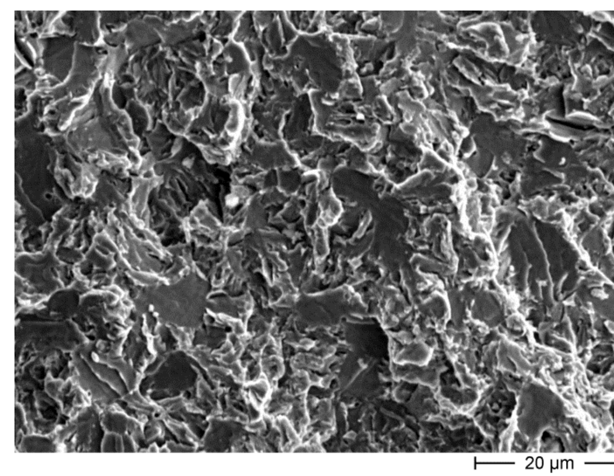




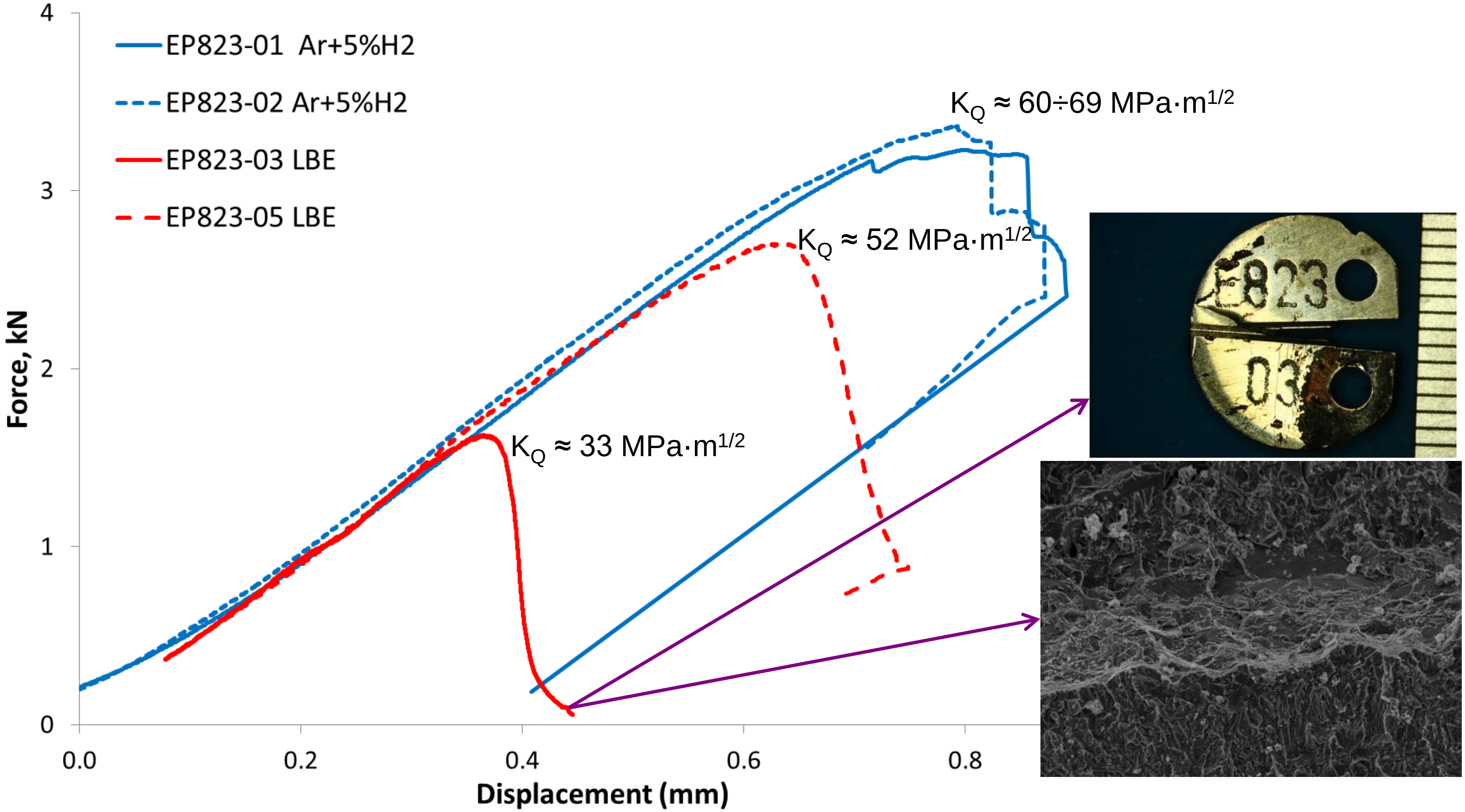
LBE ↓





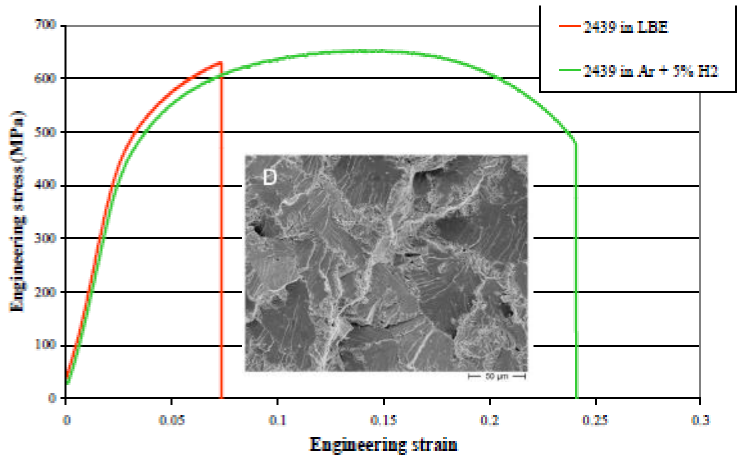


Fracture toughness test EP-823 analogue

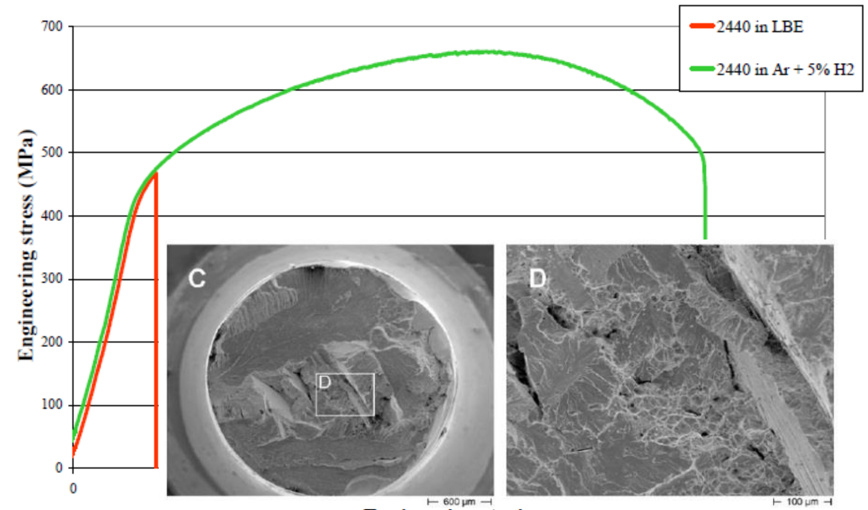


Other Si doped steels

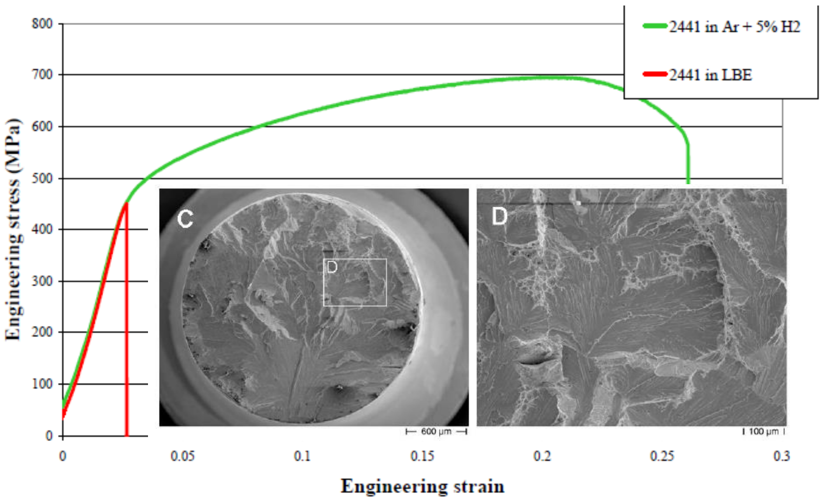
2439 steel (11.58 wt% Cr; 0.48 wt% Ni; 2.75 wt% Si)



2440 steel (13.52 wt% Cr; 0.51 wt% Ni; 4.8 wt% Si)



2441 steel (18.35 wt% Cr; 0.51 wt% Ni; 5.2 wt% Si)



350 °C
Strain rate: 5E-6
[O]: saturated

Van den Bosch, J., et al., *On the LME susceptibility of Si enriched steels.* Journal of Nuclear Materials, 2012. **429**(1–3): p. 105-112.

Materials tested at SCK•CEN for susceptibility to Liquid Metal Embrittlement in LBE by SSRT

- **T91** DEMETRA heat - screening tests completed->susceptible
  - irradiated - screening tests completed –> very susceptible
- **316L** DEMETRA heat -> screening tests completed-> not susceptible
  - Irradiated (up to 30dpa) –> not susceptible
- **1.4970** -> screening tests completed
  - solution annealed –> not susceptible
  - Cold Worked –> not susceptible
  - Cold Worked+irradiated –> not susceptible
- **CLAM** & **Si doped CLAM** –> susceptible
- **Eurofer 97 heat 2** – screening tests completed ->susceptible
- **EP-823** analog - screening tests completed – very susceptible
- **Si doped FeCr** steels->screening tests completed –>very susceptible
- **Fe10CrAl (exp. heat)** -> screening tests completed ->susceptible
- **ODS 12%Cr (KOBELCO)** -> screening tests completed ->susceptible

Investigation of LME in EU projects

- FP6 EUROTRANS (2005-2010)
  - Observations of LME in various mechanical tests
- FP7 GETMAT (2008-2013)
  - Investigation of irradiation effects
- FP7 MATTER (2011-2015)
  - Development of testing procedures guidelines
- FP7 MatISSE (2013-2017)
  - Investigation of mechanisms and development of mitigation approaches
- H2020 GEMMA (2017-2020)
  - Qualification of welds

Effects of environment for reactor structural components

- Irradiation Embrittlement
  - Reactor pressure vessel of LWR
  - Surveillance & Master curve
  - Incorporation of radiation effects in RCC-MRx and fusion SDC
- Stress Corrosion Cracking (SCC)
  - PWSCC / IGSCC / IASCC
  - Disposition curves
- Corrosion fatigue in LWR
  - Fatigue endurance with environmental factor (F_{en})
  - NUREG for new reactors and licensing renewal
  - Attempts to incorporate in ASME
- Corrosion/erosion
  - Avoidance of severe corrosion
  - Corrosion allowance

Options to handle LME for design

- To use materials, which are not susceptible to LME
  - Pro: qualification program -> demonstration of immunity
  - Con: significant reduction of candidate materials number
  - Challenges: to define "immunity"
- Incorporation of LME by reduction of mechanical properties
  - Pro: widening list of candidate materials
  - Con: extensive R&D required
  - Challenges: to define conservatism of the "reduction"
- Mitigation techniques
  - Pro: vanishing of susceptibility
  - Con: questionable visibility
  - Challenges: to define and justify mitigation strategy

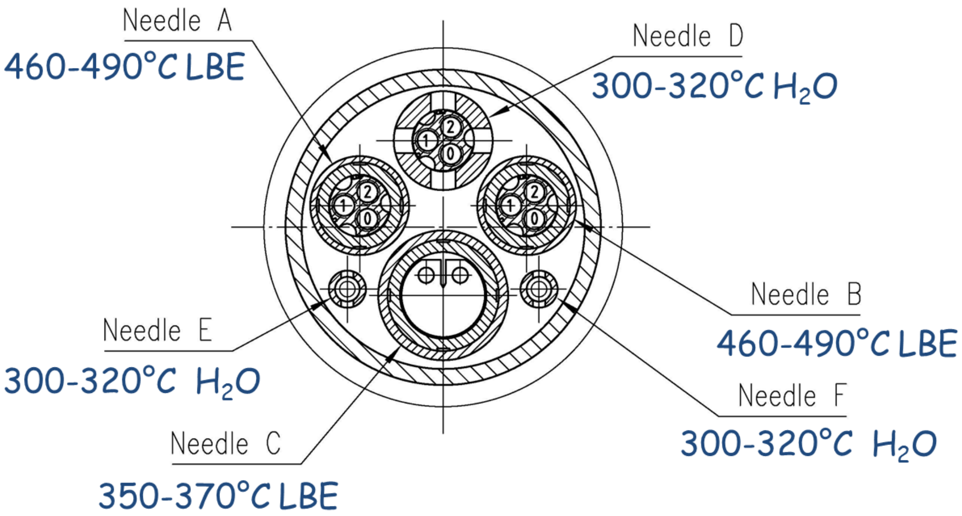
Materials degradation effects to be investigated

- Liquid Metal Corrosion (LMC)
- Liquid Metal Embrittlement (LME)
- Irradiation effects
- Synergetic effects

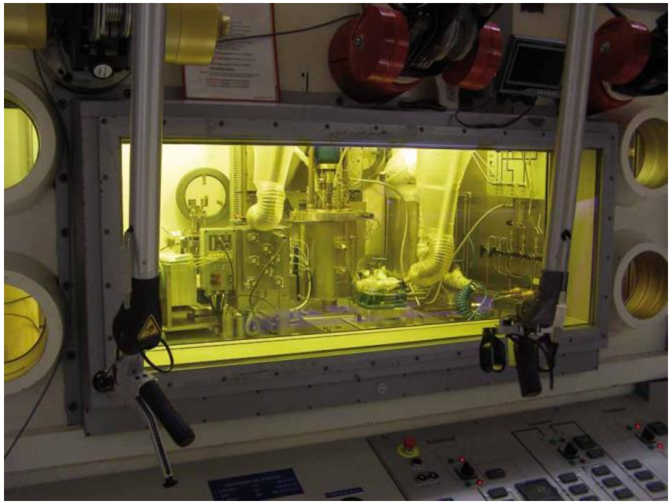
Irradiation experiments

- TWIN-ASTIR
  - Irradiation experiment in BR2 reactor (SCK•CEN, Belgium)
  - Materials: T91, 316L, High Silicon Steels, welds
  - Doses: 0, 1.5 and 2.5 dpa
  - Environment: LBE & PWR water H_2O
  - Temperatures: 300-320°C (H_2O), 350-370°C & 460-490°C (LBE)
  - Specimens: Tensile, DCT, corrosion plates
- LEXUR II
  - Irradiation experiment in BOR-60 reactor (RIAR, Russia)
  - Materials: T91, 316L, 15-15Ti, ODS (Pb)
  - Doses: 0, 6÷35 dpa
  - Environment: LBE, Pb
  - Temperatures: 350°C (LBE) & 550°C (Pb)
  - Specimens: Tensile, DCT, corrosion discs, pressurized tubes

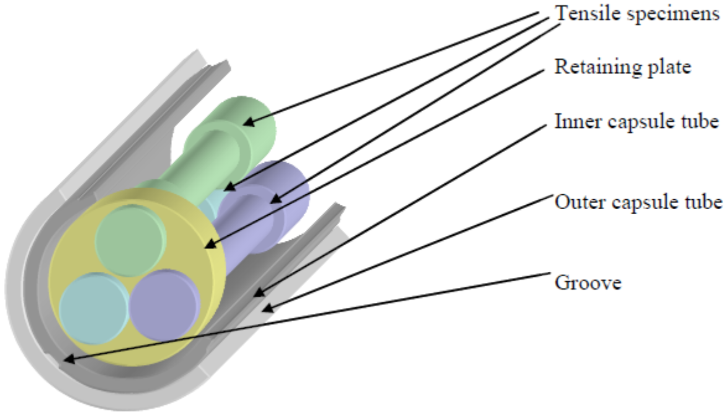
TWIN-ASTIR



Hot cell 12 & LIMETS 2

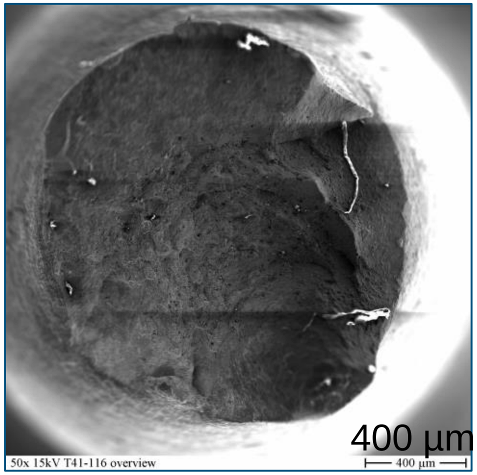


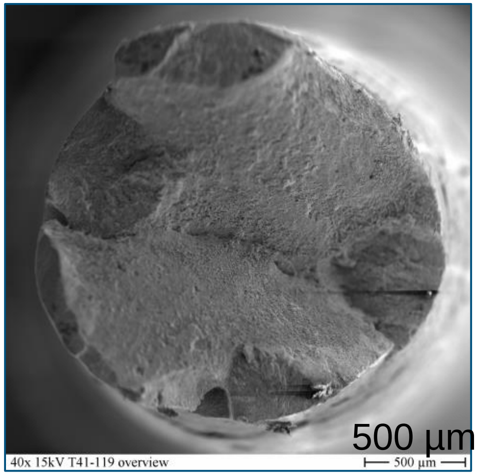
Hot cell for handling Po contaminated materials

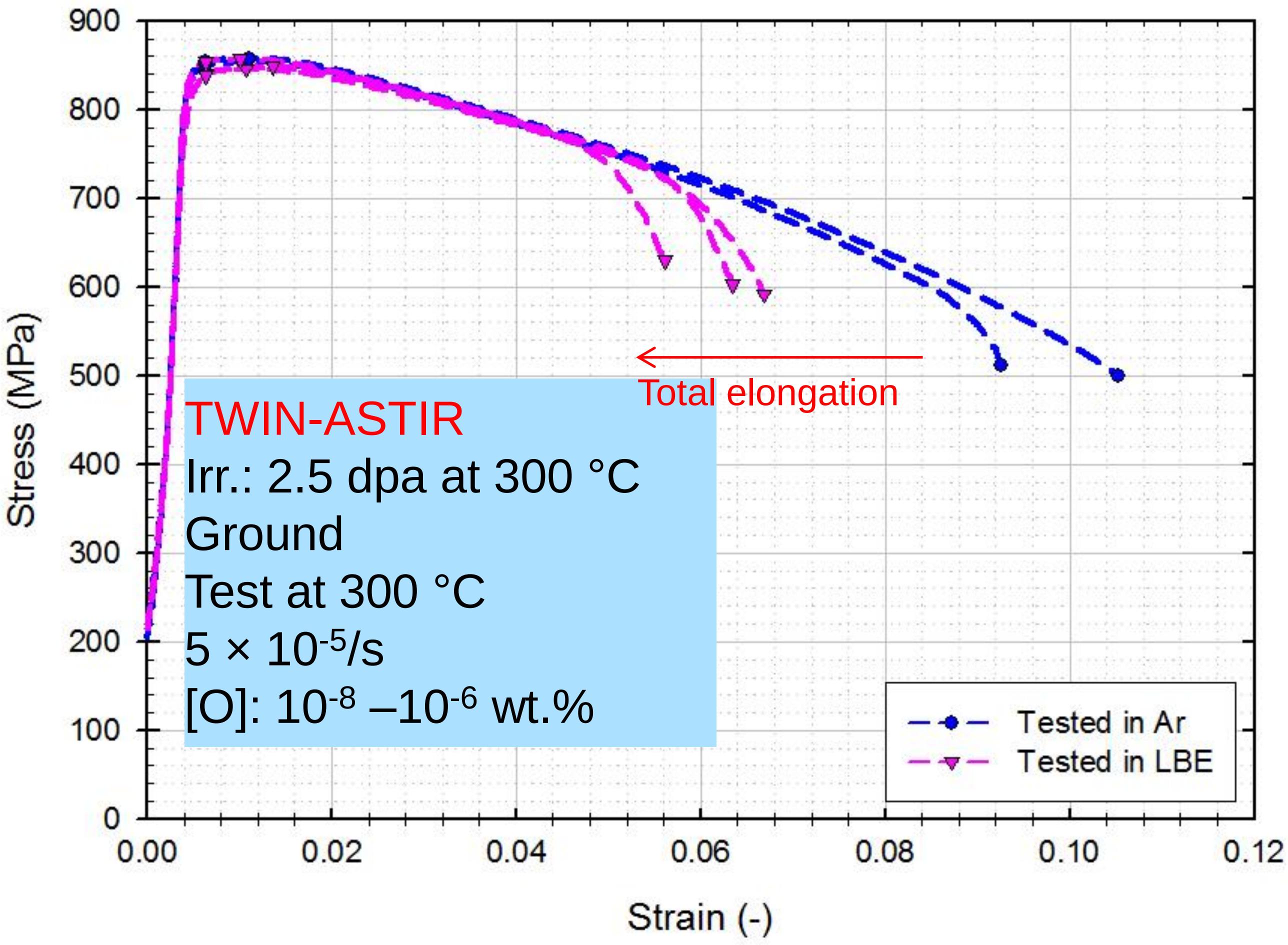


- 2006: Irradiation in BR2
- 2008: temporary license of cell 12 as α-cell
- 2008: Dismantling LBE needle A
- 2008-2009: PIE (mechanical tests)
- 2010-2011: license extension and needles E, F, D dismantling
- 2012: PIE (mechanical tests + microstructural examination)

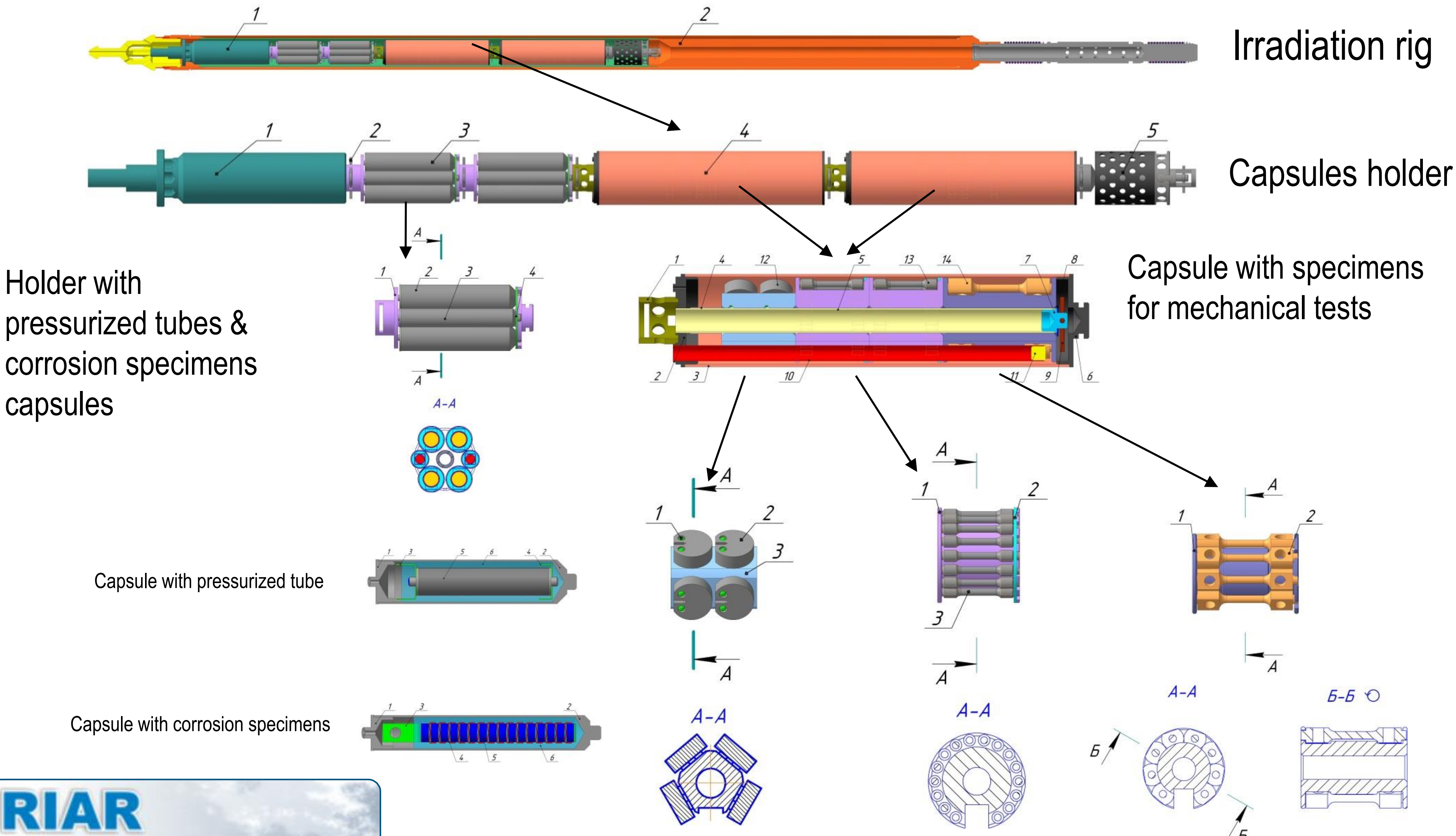
Tests of irradiated specimens: T91 : Susceptibility to LME after irradiation





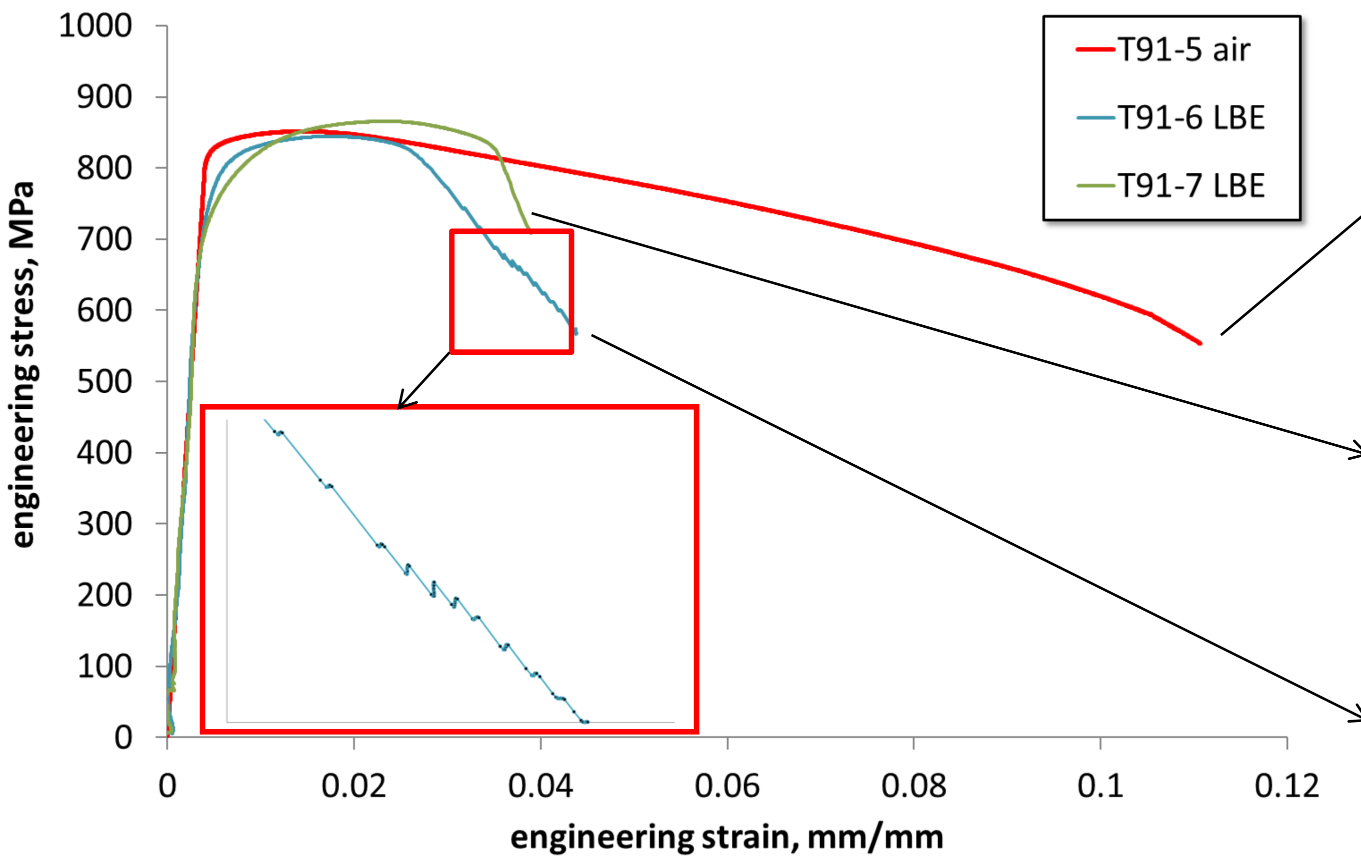


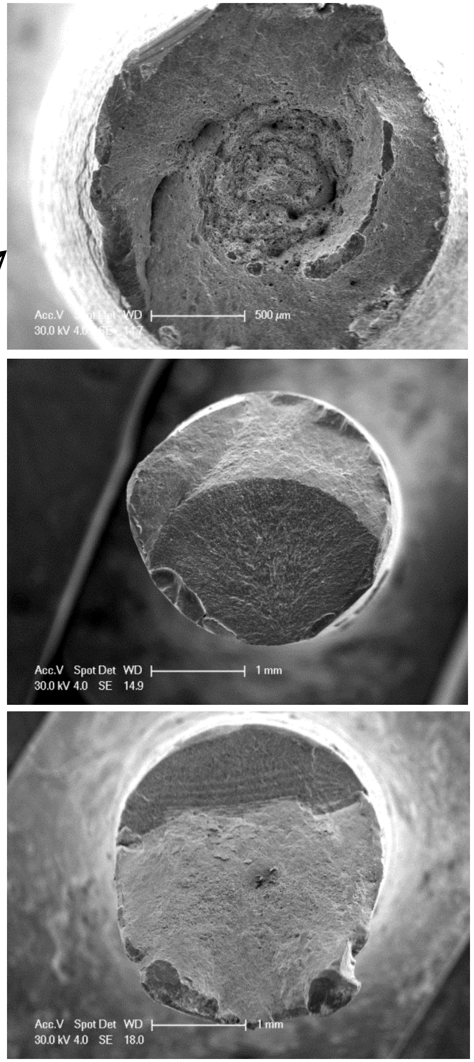
LEXUR II



RIAR
State Scientific Center -
Research Institute of Atomic Reactors

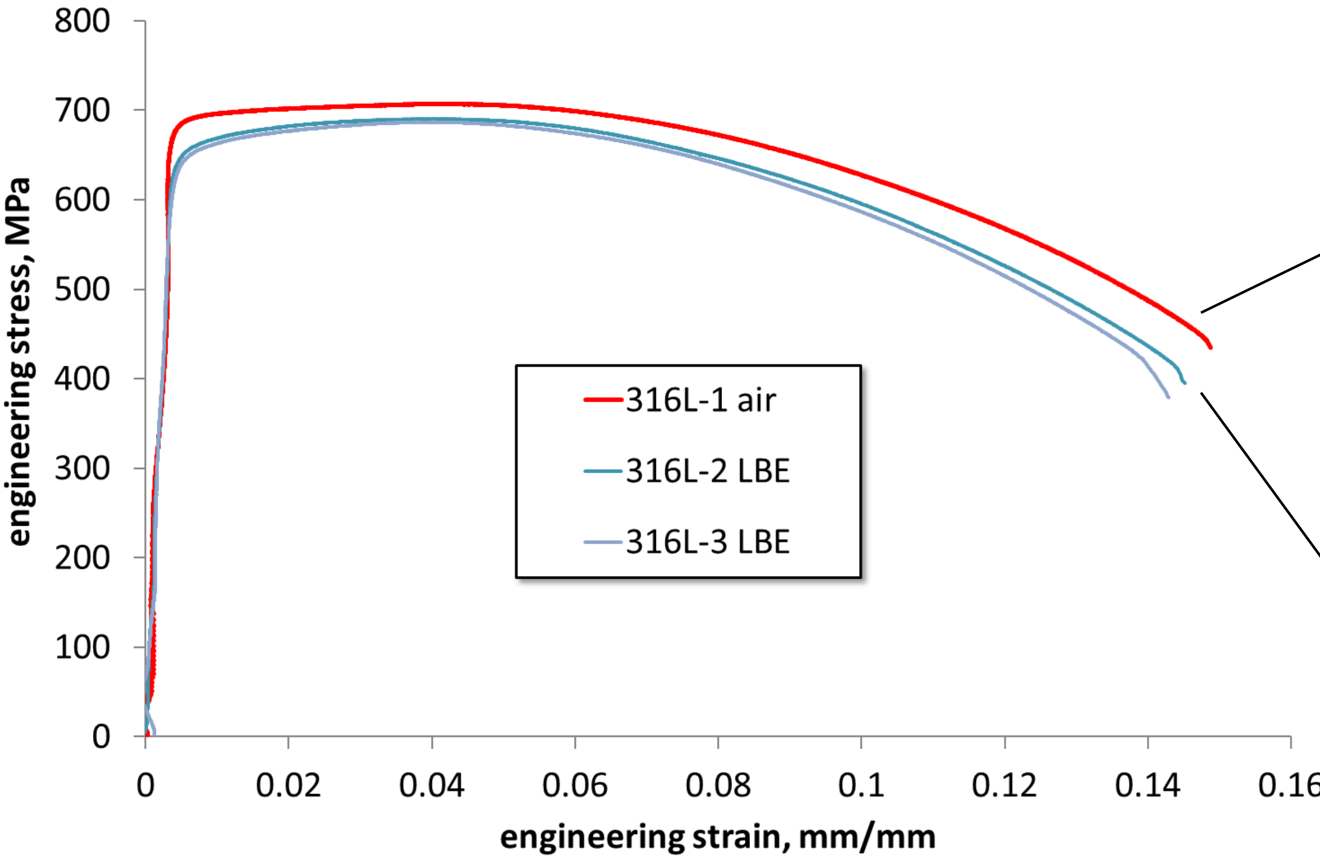
LEXUR II: LME on T91(6.1 dpa/350°C/LBE)

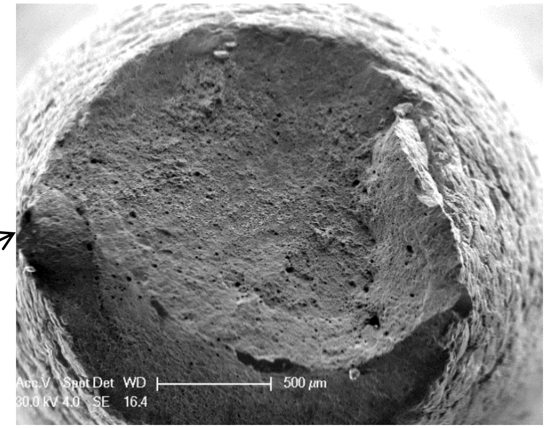


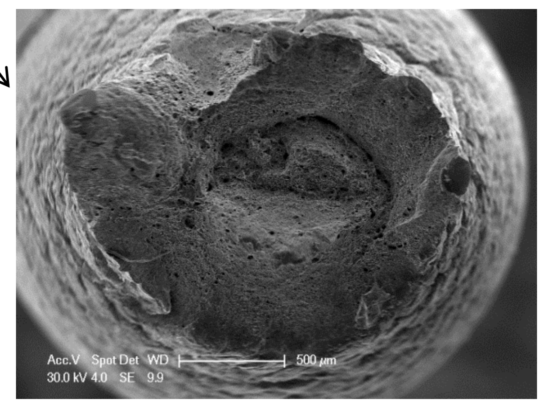


Stress-strain curves for slow strain rate tests, at **5·10^{-5}s^{-1}**, of **T91** steel irradiated in LBE at 350 °C to **6.1 dpa** in air and in **oxygen saturated LBE**, at 350 °C.

316L tensile (6.1 dpa/350°C/LBE)







Stress-strain curves for slow strain rate tests, at **$5 \cdot 10^{-5} s^{-1}$**, of **316L** steel irradiated in LBE at 350 °C to **6.1 dpa** in air and in **oxygen saturated LBE**, at 350 °C.

Summary

- Qualification of candidate materials is the key issue for the deployment of reactor systems with HLMC
- The early systems will rely on existed materials qualified for application in SFR
- Incorporation of environmental effects on material properties in the design are is the most challenging tasks for materials qualification
- These issues are in the agenda of joined European programs and prominent subject for collaboration
- MYRRHA is the fast spectrum irradiation facility for EU needs