Challenges of materials qualification for nuclear systems with heavy liquid metal coolant: Effect of LBE on materials properties

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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)
MYRRHA: accelerator driven system (ADS)

- **Accelerator**
  - (600 MeV - 4 mA proton)

- **Reactor**
  - Subcritical or Critical modes
  - 65 to 100 MWth

- **Spallation Source**
- **Fast Neutron Source**

- **Multipurpose Flexible Irradiation Facility**
Role of material research for development of MYRRHA

**Design tools:**

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

**Materials properties**

**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
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, $7.5 \times 10^{-13} < C_O \text{ (wt\%)} < 2.8 \times 10^{-8}$, 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 ($v \approx 2 \text{ m/s}$)  
   *(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

- Moderate temperatures
- Understanding of underlying mechanisms
- Active oxygen control
Solutions for LMC

- **“Short term” solution**
  - Investigation of LMC in depth
  - Database on corrosion for candidate materials including welding joints
  - 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
  - Weld overlay
  - Coatings
  - Qualification for application on functional & structural components

- **Long term solution**
  - Development of nuclear grade corrosion resistant materials
    - FeCrAl
    - Alumina Forming Austenitic Steels (AFA)
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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

Example of Liquid Metal Embrittlement

![Graph showing stress-strain curve for T91 at 350°C in Ar+5%H2 and LBE conditions.](image)
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.”

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...
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

Stress-strain curves for slow strain rate tests, at $5 \cdot 10^{-5} \text{s}^{-1}$, 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 dissolved oxygen, at 350 °C.

Effect of displacement rate on FT
Fatigue endurance

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


Investigation of susceptibility of 316L

SSRT

Fractography

Fatigue

\[ \ln N = 6.891 - 1.92 \ln (\varepsilon_a - 0.112) \]  
(Chopra and Shack, 2007)

\[ \ln N = 6.954 - 2.01 \ln (\varepsilon_a - 0.167) \]  
(ALME Code Mean Curve)
EP823 analogue: Fractured surface

EP823 analogue
350 °C
Strain rate: 5E-6 s⁻¹

Ar+5%H₂ →

LBE

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SCK•CEN
Fracture toughness test EP-823 analogue

- $K_Q \approx 33 \text{ MPa} \cdot \text{m}^{1/2}$
- $K_Q \approx 52 \text{ MPa} \cdot \text{m}^{1/2}$
- $K_Q \approx 60 \div 69 \text{ MPa} \cdot \text{m}^{1/2}$
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

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

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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

- 2006: Irradiation in BR2
- 2008: temporary license of cell 12 as α-cell
- 2008: Dismantling LBE needle A
- 2008-2009: PIE (mechanical tests)
- 2008-2009: PIE (mechanical tests + microstructural examination)
- 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**

**TWIN-ASTIR**
- Irr.: 2.5 dpa at 300 °C
- Ground Test at 300 °C
- 5 × 10^{-5}/s
- [O]: 10^{-8} – 10^{-6} wt.%

**Graph Details:**
- Stress (MPa) vs. Strain (-)
- Tested in Ar
- Tested in LBE

Total elongation
LEXUR II

Irradiation rig

Capsules holder

Holder with pressurized tubes & corrosion specimens capsules

Capsule with pressurized tube

Capsule with corrosion specimens

Capsule with specimens for mechanical tests
Stress-strain curves for slow strain rate tests, at $5 \cdot 10^{-5} \text{s}^{-1}$, 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 \(5 \times 10^{-5}\) 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.