Experimental investigation of beryllium: plans and current results within the RaDIATE collaboration

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Content

• Context of the research
• Materials, point of interest
• Microstructural investigation proton irradiation vs ion implantation
• Mechanical properties
• Conclusions
Beryllium is a promising candidate because of:

- good “nuclear” properties;
- appropriate mechanical properties
- good “thermal” properties (conductivity, specific heat, melting point);
- high oxidation resistance;
- positive experience from existing facilities
Where will Beryllium be used?

### Long-Baseline Neutrino Experiment (LBNE)

<table>
<thead>
<tr>
<th>Application</th>
<th>Operating conditions</th>
<th>Proton beam parameters</th>
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</thead>
<tbody>
<tr>
<td></td>
<td>Avg. T (°C)</td>
<td>Peak T (°C)</td>
</tr>
<tr>
<td>Beam window (vacuum to air)</td>
<td>200</td>
<td>300</td>
</tr>
<tr>
<td>Target</td>
<td>375</td>
<td>450</td>
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**Size:**
- Target: L = 950 mm, D = 15.3 mm (48 sections)
- Window: 25.4 mm diameter, 0.25 mm thick

**Environment:** elevated temperature + radiation + pulsing loads
From: Matthews (CCFE). Overview of the JET ITER Overview ITER–like Wall first results and scientific programme first programme. APS Salt Lake City, November 2011

Experience exchange with fusion community
What can we expect during irradiation?

Microstructural response:
- creation of transmutation products;

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• creation of transmutation products;
• creation and agglomeration of point defects;
• segregation (precipitation) or depletion on point defect sinks
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- creation and agglomeration of point defects;
- segregation (precipitation) or depletion on point defect sinks

Possible irradiation effects:
- reduction of fracture toughness
- irradiation induced hardening
- reduction of ductility
- reduction of thermal conductivity
### Materials

<table>
<thead>
<tr>
<th></th>
<th>PF-60</th>
<th>S-200-F</th>
<th>S-65</th>
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<tbody>
<tr>
<td></td>
<td>Max impurities, appm</td>
<td>Max impurities, appm</td>
<td>Max impurities, appm</td>
</tr>
<tr>
<td>Al</td>
<td>170</td>
<td>335</td>
<td>170</td>
</tr>
<tr>
<td>C</td>
<td>450</td>
<td>1130</td>
<td>680</td>
</tr>
<tr>
<td>Fe</td>
<td>130</td>
<td>210</td>
<td>130</td>
</tr>
<tr>
<td>Mg</td>
<td>810</td>
<td>130</td>
<td>15</td>
</tr>
<tr>
<td>O</td>
<td>2900</td>
<td>5445</td>
<td>3260</td>
</tr>
<tr>
<td>Si</td>
<td>130</td>
<td>195</td>
<td>145</td>
</tr>
<tr>
<td>Be</td>
<td>balance</td>
<td>balance</td>
<td>balance</td>
</tr>
</tbody>
</table>

Method of manufacture: vacuum hot pressing

Beryllium is of industrial purity
**How can we predict the radiation effect?**

<table>
<thead>
<tr>
<th>Investigation of the as-received Be</th>
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<tbody>
<tr>
<td><strong>Investigation of the existing proton Be window</strong></td>
</tr>
<tr>
<td>- “real” GeV proton irradiation;</td>
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<tr>
<td>- irradiated volume is big enough for microstructural investigations and micromechanical tests</td>
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<tr>
<td>But: <strong>radioactivity of the sample</strong></td>
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<tr>
<th>Simulation with ion irradiation experiments</th>
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<td>- flexibility of irradiation conditions</td>
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<tr>
<td>- observations of the evolution of the microstructure structure;</td>
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<tr>
<td>- reasonable correspondence of He/dpa ratio.</td>
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</tbody>
</table>

**Low energy in-situ irradiation:**
- easy variation of irradiation parameters;

**High-energy irradiation + PIE**
- microstructural and micromechanical tests data will be available

But: **validity of the simulation should be confirmed**
Which experimental technique should be used?

TEM: defect clusters and He bubbles, precipitates stability

APT: behaviours of impurities (precipitations, segregations at point defect sinks)

Micromechanical tests: changes of mechanical properties
Local misorientation around indents made in pure Zr measured using EBSD
From http://energy.materials.ox.ac.uk/nuclear-projects/previous-projects/hydride-cracking-in-zirconium.html

TE-56 beryllium, Chakin and Ostrovsky / JNMm 2002

Be under irradiation
Phases effect (thermal ageing data):

**Fe-Al rich precipitates can:**
- affect *ductility* and *creep strength* (Jones et al. *J. Common Met.* 1964)
- be preferential sites for *corrosion* pit initiation (*Punni and Cox, Corros. Sci.* 2010)

**Fe-Be precipitates can**

**Al and Mg can**
- form *low melting point eutectics* (*Kleykamp, JNM* 2001)

**Precipitates should be investigated**
Irradiation can produce much bigger variety of phases
**n-irradiation:**
- At low $T_{irr}$ below ~ 200°C (Chakin et al. JNM 2009) or 400°C (Gelles et al. JNM 1994): “black dots” and dislocation loops.
- At higher $T$: mainly He babbles

Irr. Be, TEM, DF, dislocation loops, $T_{irr.}=70 ^\circ C$, $F = 6 \times 10^{22} \text{ cm}^{-2}$ ($E > 0.1 \text{ MeV}$) (Chakin et al. JNM 2009)

Irr. Be, TEM, BF, He bubbles loops, $T_{irr.}=413 ^\circ C$, $F = 6.5 \times 10^{21} \text{ cm}^{-2}$ ($E > 1 \text{ MeV}$) (Klimenkov et al. JNM 2013)

S-200-F, proton irradiation (120keV, RT. $2 \times 10^{18} \text{ ions/cm}^2$), (from Kang et al. Journal of the Korean Physical Society, 63, 2013)

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<table>
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<tr>
<th>Irradiation Source</th>
<th>He gas production in Be (appm/DPA)</th>
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<tbody>
<tr>
<td>Mixed spectrum fission reactor</td>
<td>10-500</td>
</tr>
<tr>
<td>High energy proton beam</td>
<td>4000</td>
</tr>
</tbody>
</table>

**Implantation of He and H:**
- bubbles can dominate even at RT

**What can we expect from GeV protons?**
300 kW NuMI beam window
(MARS calculations of Brian Hartsell, Fermilab)

- 120GeV proton beam
- about $3 \times 10^{13}$ protons per pulse, 0.5 Hz
- $1.57 \times 10^{21}$ protons during its lifetime
- 1.1mm beam sigmas, X and Y
- $T \approx 200^\circ$C
Gaussian distribution of the beam

• Radiation damage distribution is not monotonic
The main transmutation products are He and H.

Large difference of dpa and transmutants production is likely to produce non-homogeneous changes across the surface of Be window.
300 kW NuMI beam window
(MARS calculations of Brian Hartsell, Fermilab)
• 120GeV proton beam
• $1.57 \times 10^{21}$ protons during its lifetime

Be samples annealed in contact with liquid Li. Distribution of Li concentration in beryllium along a diameter of specimens. Penetration of Li into beryllium can cause the degradation of mechanical properties. I.B. Kupriyanov et al. / Fusion Engineering and Design 2010

• The quantity of Li is not negligible (up to 500 appm in the centre)
• APT for experimental validation of MARS code
Behaviours of solid (liquid) transmutation products

- Li is not soluble in Be. Will it segregate?
- \( T_{\text{melt}}(\text{Li}) = 181^\circ\text{C} \) (for bulk lithium). Can we expect the creation of liquid phase in the window?

**Solubility of Li in Be:** 130 appm at 700°C and 40 appm at 600°C (from Kupriyanov et al. / Fus. Eng. and Des. 2010)

*Example*

F82H, irradiation 0.5 Gev protons, 350°C, 20 dpa, 370 appm of Ca created
Kuksenko et al. / JNM 2014
Nano-hardness measurements:
• to find the Gaussian peak
• to estimate the irradiation effect

Local microstructural investigations

, D.Armstrong. University of Oxford
FIB lift-out

- superpose the microstructural data with the dpa, appm and hardness data
- minimize the activity of samples
- minimize the toxicity of samples
We need to know the evolution of radiation effects over the time

Collaboration with HiRadMat project (poster of Kavin AMMIGAN)

Ion irradiation experiments
Microscope and Ion Accelerator for Materials Investigations facility (MIAMI) University of Huddersfield, UK (collaboration with Prof. S E Donnelly)

**Ions:** He+

**Beam energy:** ~ 10keV => peak of damage in the middle of TEM foil (SRIM)

**Dose:** up to 1 dpa

**Temperature:** 200°C (300°C, 600°C)

From http://www.hud.ac.uk/research/researchcentres/emma/miami/
In-situ observations of the evolution of the microstructure
- evolution of number density and size of dislocation loops and/or He;
- estimation of mobility of point defect clusters
- Burgers vector and loops nature determination*

But: effect of the surface

Irradiation of APT tips?

Fe-9Cr alloy, 150keV Fe⁺ ions irradiation, 300°C
Surrey Ion Beam Centre, UK
(collaboration with Prof. R.Gwilliam)

Ions: He+
Maximum beam energy: 2 MeV => 7.5µm implantation depth (SRIM)
Dose: up to 1 dpa
Temperature: 200ºC (100ºC, 400ºC)

Micromechanical tests

PT sample

8 x 8 µm³
Why use micro-cantilever testing?

- Useful where only small samples are available (implanted layer)
- Need for a sample design that can be machined in surface of bulk samples
- Geometry that can be manufactured quickly and reproducibly
Conclusions

Experimental database of the high-energy proton irradiation effects in Be is very limited.

Experimental investigation of beryllium within Radiate project should cover 3 main goals:

• characterization of existing GeV proton irradiated Be samples;
• simulation of proton irradiation effect by ion implantation experiments;
• prediction of the microstructural evolution for new irradiation conditions.
Thank you for your attention!
What do we know?

**enhancement of phase transformation**

\[ D^* = \alpha_V D_V C_V^* + \alpha_x D_x C_x \]

\[ C_V^* > C_V^T \]

- \(X\) - self interstitial atom; clusters of point defects

**agglomeration of point defects**

- **self-interstitials**
  - clusters;
  - dislocation loops.

- **vacancies**
  - voids;
  - dislocation loops.

**segregation (precipitation) or depletion on point defect sinks**

- **voids (He bubbles)**
- **grain boundary**
- **dislocation line**
- **precipitates**
- **dislocation loop**

\(V\)- vacancy; \(I\) - interstitial

**inverse Kirkendall effect**

If \(D_B^V < D_A^V\) → depletion of A atoms

**drag effects**

- B-V complexes or B-I complexes

Segregation of B atoms
Helium has complex effects on both yield and fracture properties of tungsten. Differences between results for micro-cantilevers and nanoindentation show the difficulty of relying on one type of test.
Micro Cantilevers Before Testing
Precipitates **Fe and Al rich precipitates** may affect **ductility** and **creep strength** (A.W. Jones, R.T. Weiner, J. Common Met. 6 (1964) 266.)

Grain, twin and sub-grain boundaries and dislocations can be the preferential places for precipitation of **Fe-rich phases** during ageing of Be-0.25%Fe. Dislocation can locked by precipitates leading to the increase of **hardness** (S. Morozumi, N. Tsuno, S. Koda, Trans. Jpn. Inst. Met. 10 (1969) 64.)

Intermetallic **Fe/Al/Be inclusions** are the preferential sites for **corrosion** pit initiation, some corrosion pits had also initiated at elemental **Si** and **carbide** inclusions. (J.S. Punni, M.J. Cox, Corros. Sci. 52 (2010) 2535)

Al and Mg can form **low melting point eutectics** in Be, that might influence the mechanical behaviour of Be.

\[ e^{-((x^2)/3+(y^2)/3)} \]
Nanoindentation mechanical probe which allows local hardness and modulus to be measured.

Micrometre

Hardness of W5Ta after self-ion irradiation

Will be used for high-energy ion irradiation samples and NuMi window (if not too “hot”)