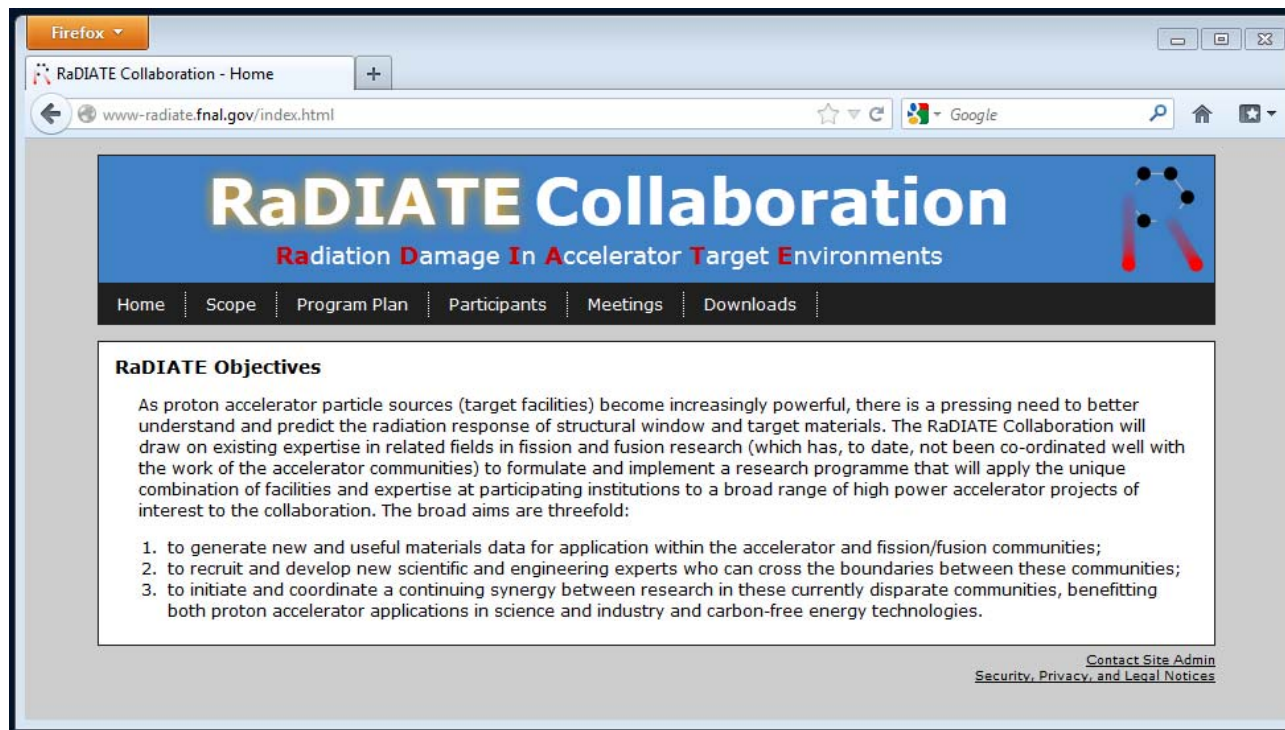


RaDIATE Collaboration

K. Ammigan, R.B. Jones *et al.*

Proton Accelerators for Science and Innovation
2nd Annual Meeting
RAL, UK
19 May 2013

RaDIATE Website



Web address: : <http://www-radiate.fnal.gov/index.html>

Listserv address: radiate@listserv.fnal.gov

- Scope and plan of the collaboration
- Presentations and minutes from monthly meetings (5)
- Documents and reports on radiation damage studies
- Irradiated materials table and target parameter space

Stage 1 progress: Exploratory/Development

Stage 1 objective: develop specific research activities to meet program goal!

- Literature review of material radiation damage studies
Graphite, Beryllium and Tungsten
- Post-doc material of study choice
- Post-doc recruitment
- Irradiated specimens and target parameter space
- Identify existing facilities testing capabilities



Radiation damage literature review

Graphite

- Barry Mardsen and Graham Hall (University of Manchester)
- Radiation damage in nuclear graphite
- Analyzed potential concerns for target environments

Beryllium

- Barry Jones (Oxford University team) to provide an update on Be work
- Lack of radiation damage data on fatigue or irradiation creep

Tungsten

- Barry Jones (Oxford University team) starting to look at Tungsten data in more detail



Material of choice for post-doc studies

Beryllium

- If RaDIATE does not undertake Be radiation damage study, it will not happen anytime soon!
- Research programs on graphite and tungsten already ongoing for nuclear applications
- Broad application as windows in all high intensity machines
- Some experience with Be from fusion research at Culham Center for Fusion Energy (CCFE)
- LBNE funding significant portion of post-doc cost – low Z neutrino target material suitable for LBNE is desired



Post-doc recruitment

- Post-doc to be based at the Materials for Fusion and Fission Power (MFFP) group at Oxford University under Steve Roberts
 - anticipated work at RaDIATE US participating institutions

- Job description for post-doc Be studies finalized

- PO for post-doc costs in process

- Advertising very soon!



Irradiated specimens and parameter space

- Updated list of available irradiated specimens
 - Be, W, C
 - Irradiation environment and proton fluence
 - 0.16 – 400 GeV, 60 - 900 °C, $5e20 - 5e22$ p/cm²
 - Estimated DPA damage and gas production
 - 0.1 – 9 DPA, ~ 300 appm/DPA He, ~ 1000 appm/DPA H

- Parameter space for RaDIATE program
 - Targets/windows of future accelerator facilities
 - Expected operating conditions and beam parameters
 - 0.8 – 120 GeV, 300 - 1600 °C, 0.1-20 DPA/yr
 - Potential tests/studies required

<http://www-radiate.fnal.gov/downloads.html>



Potential new collaborators

- ❑ FRIB/MSU MatX initiative – Georg Bollen, Frederique Pellemoine
 - Radiation damage from ‘swift ions’ on
 - Graphite target
 - Titanium beam dump vessel
 - Diamond detectors

- ❑ LANL – Stuart Maloy has expressed interest and is on the RaDIATE mailing list

- ❑ ESS Target Facility
 - Tungsten target
 - Beam window



Other news...

☐ MOU status

- **Approved!**
- Current collaborators: FNAL, STFC, PNNL, BNL, Oxford

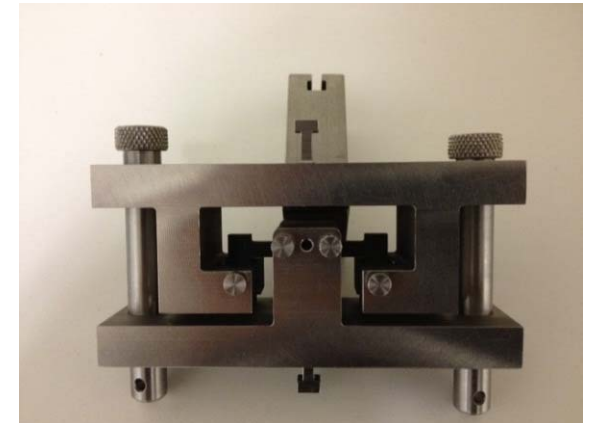
☐ NNUF update from Steve Roberts

- Currently purchasing equipment
- Potentially accept activated materials for tests in about 2 years

☐ BNL BLIP graphite studies

- HEPA filters and hot cell ready
- Flexural tests on irradiated 3D C/C composites

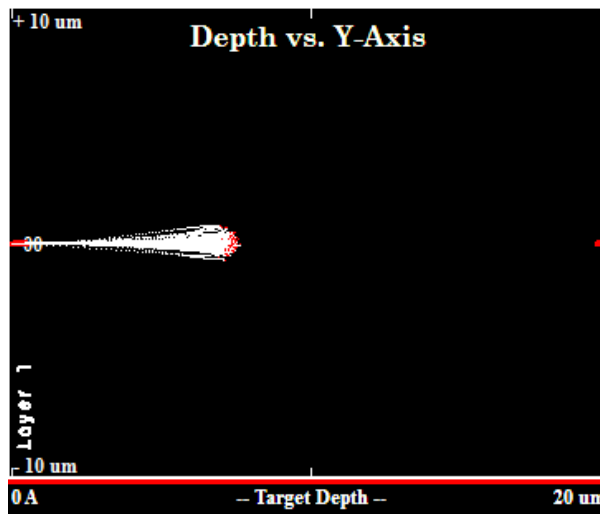
☐ Interim report for Stage 1 complete



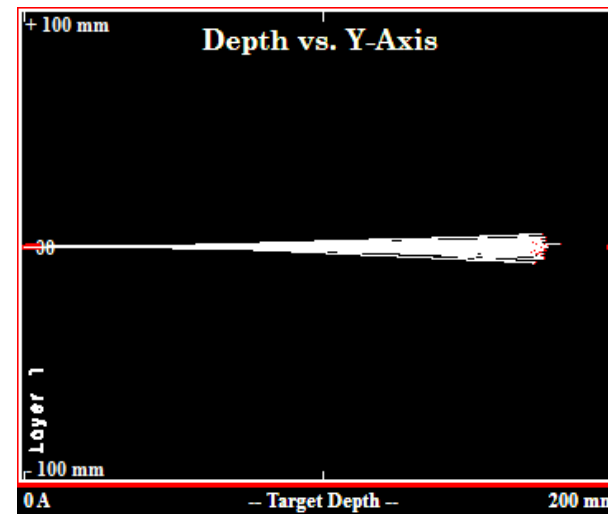
Next steps...

- ❑ Final report for Stage 1 – Summer 2013
- ❑ Recruit post-doc by Fall 2013
- ❑ Evaluate available irradiated specimens and new irradiation testing needs – HE protons vs. LE ions

2 MeV He ions into Be
~ 7 μm penetration

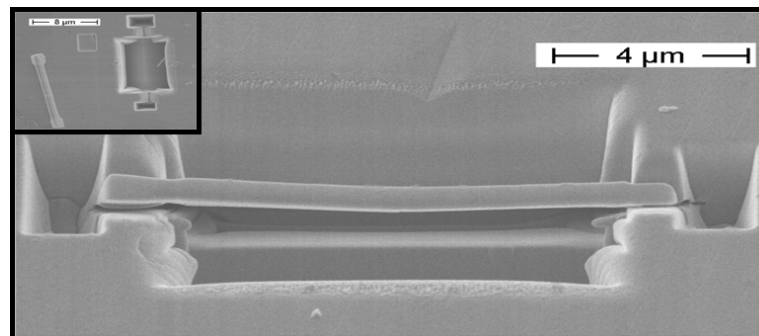
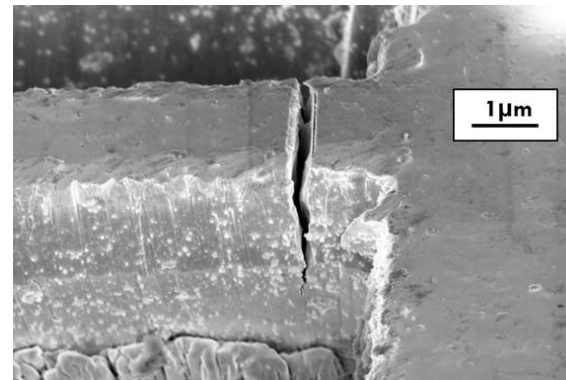
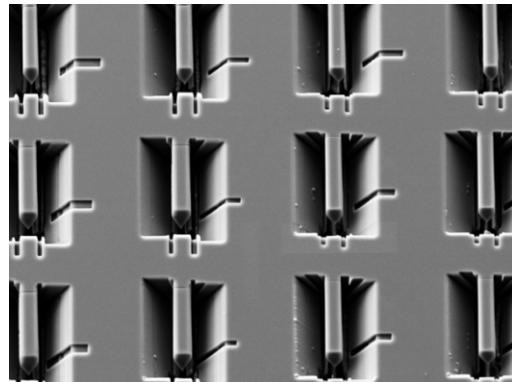


200 MeV protons into Be
~ 180 mm penetration



Next steps...

- ❑ Evaluate Post Irradiation Examination (PIE) techniques to start initial tests on irradiated Be specimens
- ❑ Micromechanics on Be (MFFP group – Oxford)
 - Micro-properties to bulk properties



Opportunities for future collaboration

- ❑ Coordinate with FRIB/MSU on MatX initiative
- ❑ Graphite: BNL BLIP
- ❑ Tungsten: ISIS/RAL
- ❑ Open to other collaborators
 - Contact: P. Hurh, C. Densham



Beryllium for proton accelerator windows and targets

By

R B Jones

Irradiation experience on Be

- Design conditions proposed for Be proton accelerator windows and targets.
- Accelerator operating experience with Be.
- Survey of neutron irradiation experience with Be.
- Other factors of importance for Be usage.

Proposed window/target designs

LBNE 700kW, 120GeV, 1Hz, $\sigma_{\text{rms}} = 1.3\text{mm}$ Window/target

<i>Cyclic T °C</i>	<i>Dose dpa</i>	<i>Gas</i>
200/300	0.15/year total	He 1330 appm/year
Av/peak	@7e-4 to 7e-9dpa.s ⁻¹	He/dpa = 8867
	Maximum/average	H data not available

LBNE 2.3MW 120GeV, 1Hz, $\sigma_{\text{rms}} = 1.3\text{mm}$ Target only

<i>Cyclic T °C</i>	<i>Dose dpa</i>	<i>Gas</i>
350/550	0.5/year total	He data not available
Av/peak	@2.5e-3 to 2.5e-8 dpa.s ⁻¹	H data not available
	Maximum/average	

H (Tritium) generation and release needs consideration

Be windows operate in air/vacuum conditions

Accelerator irradiation experience

- Various Be windows, targets & test pieces are available.
- Average operating temps 40° to 200°C, max peak 250°C.
- Windows in air/vacuum, targets in air, test pieces in water.
- Pulsed operation 0.5 - 7.5Hz (or a more complex cycle).
- Beams 0.2 - 400 GeV, 0.19 - 1mm spot sizes.
- Peak proton fluences mostly $1.4e21$ to $3.7e24$ p/cm².
- Peak dose (when quoted) 0.1 - 8.5 dpa.
- No gas production values available.
- Two CERN CNGS windows failed after $0.49 - 1.4e20$ POT.
- Maximum life 9 and 30 years for another window and target respectively.
- S65, PF-60 and S200F Be grades represented.
- Post-irradiation examination needed for causes of failure.

Data from neutron irradiations

Irradiations cover 43 - 600°C, doses from <1 - 52 dpa (Be) at dose rates of 0.2 - 9.9×10^{-7} dpa/s with He contents up to 22,500appm at He/dpa ratios of 50 - 420. Extent of data not equally populated for all phenomena investigated.

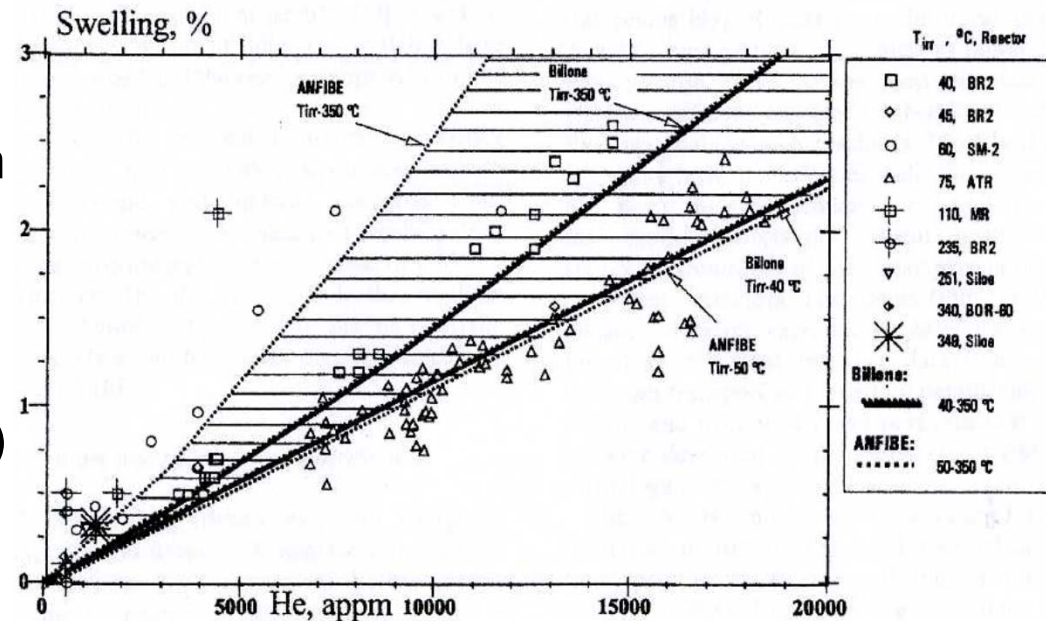
Relative to accelerators neutron exposures are at much lower He/dpa ratios and constant dose rates and temperatures. Accelerators have cyclic dose rates and temperatures

Topics investigated (* more details given below)

- *Helium and hydrogen production; bubble swelling
- Irradiation growth
- Irradiation hardening, irradiation embrittlement
- *Fracture toughness
- *Thermal creep, irradiation creep and stress relaxation
- Cyclic stressing
- *Thermal conductivity
- Irradiation induced changes to other physical properties
- Oxidation
- Corrosion

Helium gas and swelling

- Helium arises from ${}^9\text{Be}(n, 2n){}^4\text{He}$.
- Large amounts are generated.
- He bubbles give swelling after irradiation at $\geq 200^\circ\text{C}$.
- No bubbles $\leq 200^\circ\text{C}$; He accommodated in the Be lattice (solid swelling).
- Figure shows %swelling vs He appm (C) for both regimes.



$$\%V/V_0 = 1.15e-4 C [1 + 9.49e-5 C^{0.5} T^{1.5} \exp(-3940/T)], \quad (T \text{ in } ^\circ\text{K}, \text{Billone's equ'n}).$$

However – only a few data are from density measurements; length changes often used.

He content often calculated, not measured.

Older grades of Be show more swelling.

- For He level in LBNE (1330appm/year) swelling is $\leq 0.5\%/year$.
- Tritium generated by neutrons but in small amounts ($\sim 1\%$ of He). This may diffuse to bubbles or escape at surfaces.

Thermal and irradiation creep of Be

- Thermal creep important $>0.5T_m$ ($>504^\circ\text{C}$)
i.e. at the hot end of Be accelerator usage.
- Wide variations found due to impurities (Al, Mg, Si, Fe), porosity and %BeO.
- Recent data shows steady creep rate (sec^{-1})

$$\dot{\epsilon} = A (1 - p^{2/3})^{-2.43} \sigma^{2.43} \exp(-19470/T)$$

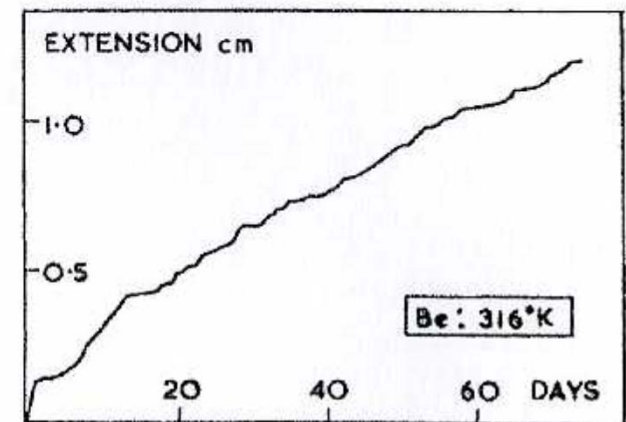
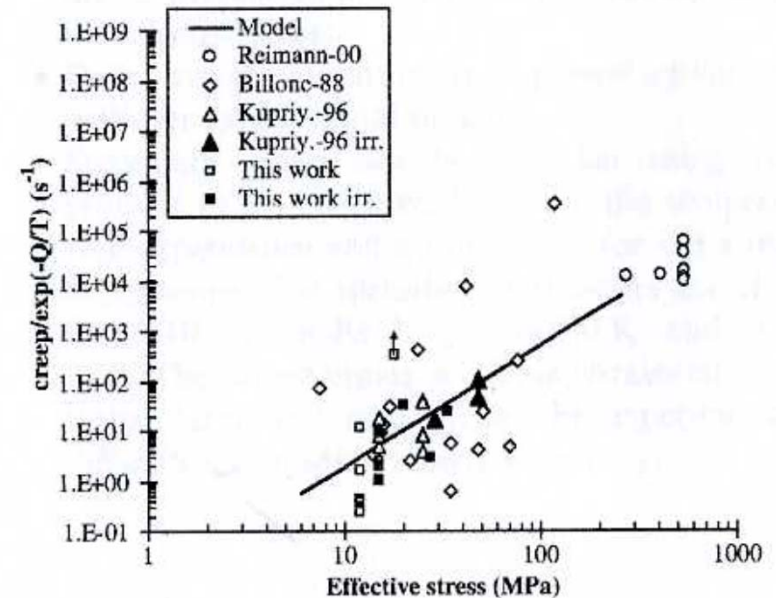
with $A = 7.21 \times 10^{-3}$ and σ in MPa. This relation incorporates post-irradiation creep for swollen Be (porosity p).

- Concurrent irradiation enhances creep. The lower figure shows a loaded Be helical spring deforming at 43°C in a MTR (only one test).
- Derived steady state irradiation creep is

$$\dot{\epsilon}_i = 3.2 \times 10^{-6} (1 - p^{2/3})^{-1} (\Phi) \sigma$$

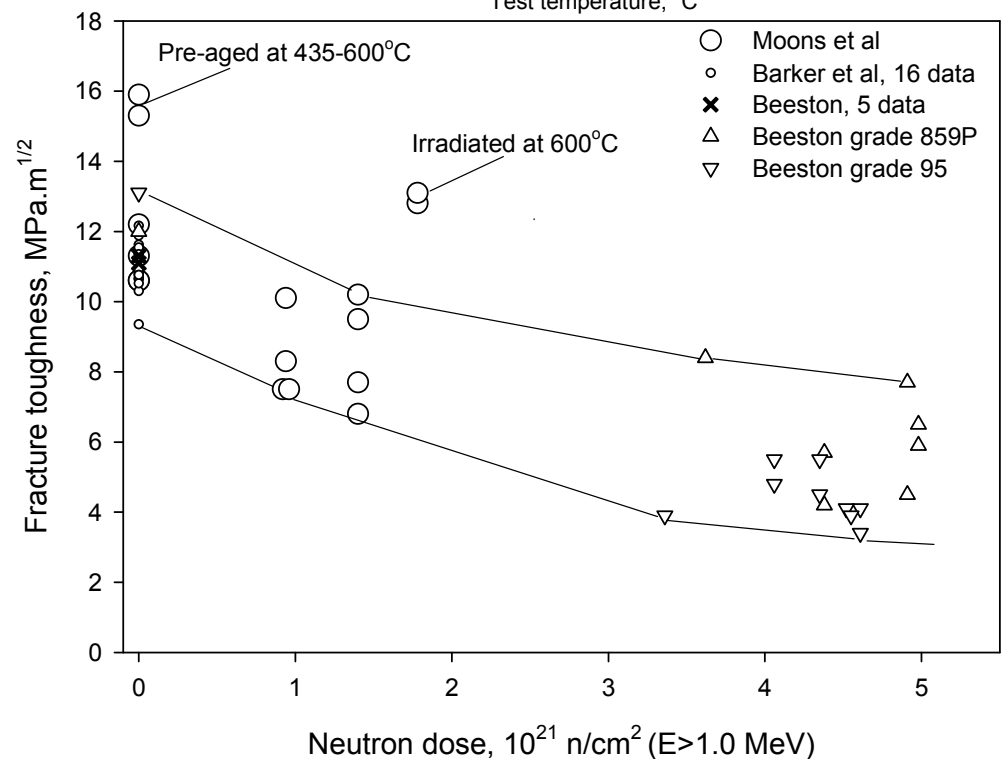
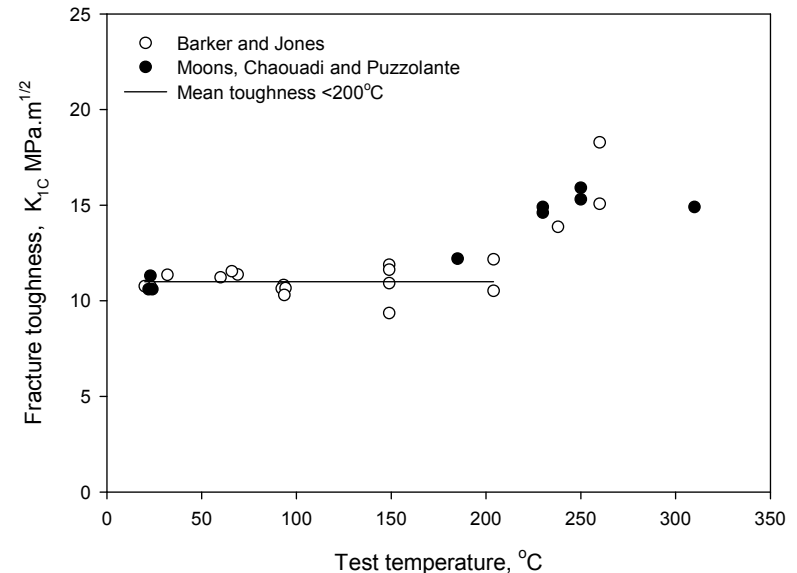
with Φ in dpa/s and σ in MPa. No primary creep or any later creep from irradiation-induced void or bubble swelling is included

- Stress relaxation can be estimated from the primary and steady creep data.



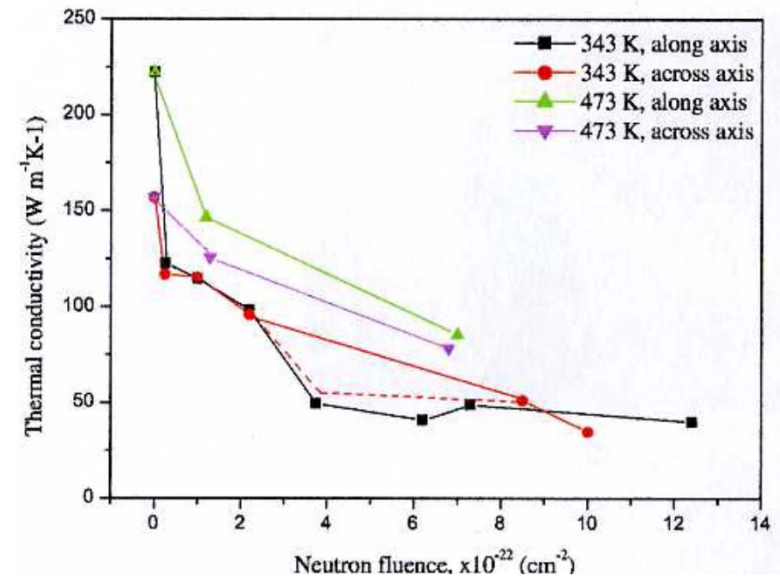
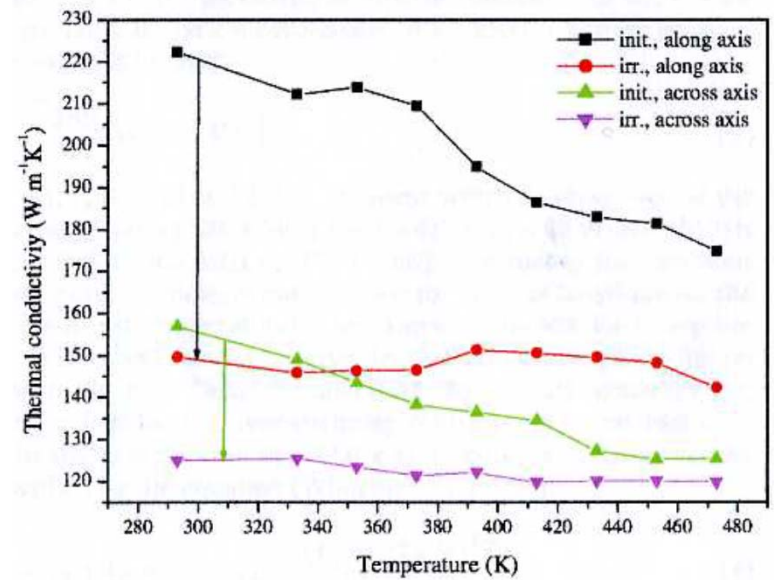
Fracture toughness of Be

- Differing grades of Be (CIP-HIP, VHP and HIP) with 0.5% to 1.6% BeO content were tested.
- All unirradiated Be grades had the same toughness of 11 MPa.m^{1/2} at temperatures <200°C. Toughness increased above ~200°C.
- Irradiations in BR-2 at 200, 230 & 350°C to 0.94 to 1.78 x 10²¹n/cm² (E>1MeV & He/dpa ratio of 290) degraded toughness (Moons et al).
- Be exposed in ATR at 66°C to 3.4 - 5.0 x 10²¹n/cm²(E>1MeV) also reduced toughness (Beeston).
- Higher temperature-aged or 600°C-irradiated Be exhibited better toughness levels before and after irradiation.
- No toughness data at high He levels.



Irradiation and Be thermal conductivity (κ)

- Irradiation effects on κ examined in Be of differing textures, grain sizes and impurity levels (mainly the level of BeO).
- Irradiations were at 343K and 473K, doses of 2 – 58 dpa and He 840 – 20,600 appm (1dpa $\sim 0.25 \times 10^{22}$ n/cm² ($E > 0.1$ MeV)).
- κ varies with test temperature and axial orientation (texture). After irradiation at 473K (see top diagram) κ is much reduced, the test temp dependence is lost but texture effects remain.
- The dose-induced reduction in κ after 673K irradiation is much smaller (only 10-20%).
- The most rapid irradiation-induced reduction in κ occurs at the lower doses and irradiation temperatures (lower diagram).
- Prismatic interstitial loops (20-80nm), basal vacancy loops (40-500nm) and He bubbles (4nm) occur (no bubbles below 473K). All loops absent at 673K, only flat plate-like pores seen.
- The reduction in κ at low irradiation temps is mainly due to radiation-induced dislocation loop formation with a contribution from He generation and small He clusters.
- No quantitative analysis has been fully developed relating the type and morphology of the defects present to the observed reductions in κ .
- Need further work at low doses at 473K and on the relative κ effectiveness of transmutation gas.



Other issues

- Irradiation growth in Be has been measured but its magnitude has not been satisfactorily separated from the effects of gas swelling.
- Evidence is available on the release of He and H (T) from Be during heating. He release is not significant below 600°C.
- Many data exist on irradiation hardening and embrittlement in Be. These effects have their maximum effect at 400°C.
- No information has been found on the cyclic stressing or creep/fatigue response of irradiated Be.
- Predictive equations are available for the physical properties of Be and the influences of temperature and porosity (e.g. density, Young's modulus, coefficient of thermal expansion).
- Evidence exists for enhanced oxidation of Be under He ion irradiation at ambient temperature. However the significance of this is tempered by the good practical performance of proton-irradiated Be windows in air.
- Long term neutron irradiation increases the corrosion of Be in cooling water.
- Be is not easy to fabricate using conventional techniques which yield textured products. Powder metallurgical methods (HIP) are favoured and can produce products with randomised grain orientations.
- Be is toxic and requires specialised handling (glove boxes, adequate ventilation etc).

Conclusions

- Post-irradiation examination of both failed and unfailed Be windows/targets is required to determine what factors affect longevity under proton irradiation.
- More data required on He and H generation by protons in Be window/targets.
- Determinations of thermal conductivity effects are needed at low irradiation doses and window/target temperatures. Obtain better definition of the κ -effectiveness of He bubbles.
- More Be irradiation creep data needed for estimating stress relaxation of thermal and mechanical stresses.
- Investigation required of pulsed irradiation doses on irradiation creep.
- Experiments needed on cyclic stressing and creep/fatigue of Be both after and under irradiation.
- Need to explore the fracture toughness response of Be at irradiation temperatures between 250 and 600°C. Determine whether toughness is affected by differing distributions of He bubbles.