The Effect of Neutron Irradiation on Candidate First Wall and Diverter Materials

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ITER Cutaway





Vertical Target

PFC : either W or Carbon Fiber Composite Heat Sink : CuCrZr alloy Structure : 316 LN(I)-G steel alloy

<u>Dome</u>

PFC : W Heat Sink : CuCrZr alloy. Structure : 316 LN(I)-G steel alloy

<u>Tile</u>

Pure Beryllium

Neutron Cascade Damage and Defect Evolution

neutroncopper collision

Before we proceed.....a definition..... DPA = displacement per atom Typical reactor application = 1-10 DPA / year 1 DPA Ceramic ~ 1 x 10²⁵ n/m² E>0.1 MeV



Lattice Displacement Energies

- Graphite : 25 eV
- **Be** 25 eV
- Iron: 40 eV

Tungsten : 80 eV

A 100 keV imparted energy Will produce thousands of displaced atoms !



Five Evils of Radiation Damage (in Metals)

- Radiation hardening & embrittlement (<0.4 T_M, >0.1 dpa)
- Phase instabilities from radiation-induced precipitation (0.3-0.6 T_M , >10 dpa)
 - High temperature He embrittlement (>0.5 T_M, >10 dpa)
- Volumetric swelling from void formation (0.3-0.6 T_M, >10 dpa)
- Irradiation creep (<0.45 T_M, >10 dpa)









Evils of Radiation Damage (in Ceramics...)

Graphite Sublimes at > 3000 K

• Point Defect Swelling and Amorphization (<0.3 T_M, < 5 dpa)

• Radiation Induced Thermal Conductivity Degradation (<0.4 T_M, < 5 dpa)

- Volumetric swelling from void formation (0.4-0.6 T_M, >4 dpa)
- Irradiation creep (<0.45 T_M, >10 dpa)









Operating Range, Irradiated Structural Materials



Copper for Nuclear Application

• Copper has been used extensively in tokamak application for heat sink application.

• Annealed, pure copper, is extremely weak at both room and elevated temperature. Solid solution, precipitation hardened, or dispersion strengthened copper alloys are used to increase strength.

• Neutron irradiation-produced defects cause significant embrittlement in copper.



face center cubic 1357 K Melting Point



Defect Production and Annihilation in Irradiated Materials

vacancy • interstitial

-strong function of material, time, and temperature...

Condensation of Cascade in FCC Copper

A large non-perfect SFT and several SIA clusters/loops are formed

Defect cluster microstructure in Cu irradiated to 1 dpa at low temperature (~90°C)



Dislocation dynamics and in-situ TEM of dislocation SFT interaction



Yuri Osetskii, ORNL

Yoshi Matsukawa, ORNL (now U. mich)



Irradiation Hardening and Embrittlement

• Irradiation produced defects serve as "road blocks" to the dislocation motion required for deformation (plasticity.)

• Defects can be formed either directly within the cascade or can develop (mature) upon diffusion of interstitial/vacancy species following cascade:

SFT in Cu "in cascade"



• Immobile defects formed directly within the cascade are extremely troublesome, as we have no "metallurgically fix".

- typically, higher Z metals have higher fraction of "in cascade defects"

• However, migrating defects (or transmutants such as He) can be dealt with by various means (alloying, dispersion strengthening, etc.)

CuCrZr for ITER Application

• Copper alloys have significantly higher strength than pure copper, and still retain good conductivity

•Three copper alloys were considered:

-Precipitation-hardened CuCrZr

-Precipitation-hardened CuNiBe

-Oxide-dispersion-strengthened (GlidCop AI-25)



•CuCrZr was selected due to its high fracture toughness, availability and low cost.

•ITER grade CuCrZr: -Cu - (0.60-0.90%)Cr - (0.07-0.15%) Zr

CuCrZr -vs- Pure Copper

 Precipitation hardening of CuCrZr alloy extends useful life of copper. Embrittlement and "plastic instability" stills occurs, but at a much higher level of dose







Meimei Li, M.A. Sokolov, and S.J. Zinkle, (2009) & Edwards et al (2005).

Channels formed in CuCuZr similar to the case in OFHC Cu, except that CuCrZr channels were not free of dislocations and defects

CuCrZr Tensile Properties Under Irradiation



CuCrZr loses its uniform elongation rapidly under irradiation

"Elevated" Temperature Irradiation Results for CuCrZr



• for irradiation in the 0.3-0.6 Tm range, copper embrittlement becomes less of an issue, however, alloy has little strength at these temperatures.

Effect of Neutron Irradiation on Fracture Toughness

- Neutron irradiation effect on fracture toughness is small; Fracture toughness was reduced slightly after irradiation.
- No correlation between tensile ductility loss and fracture toughness



Biased Summary Comments on Copper Alloys

• Copper and copper alloys has been very well studied, both the fundamentals of the alloys and their irradiation effects. Improvements in both non-irradiated and as-irradiated performance will be incremental.

• Copper alloys are inherently limited in elevated temperature strength. For this reason they may not be attractive alloys beyond ITER.

Tungsten for Nuclear Application

- Currently, tungsten is utilized as a plasma facing "tile" material with only moderate structural requirements. For ITER the requirement for ductility and irradiation resistance is limited.
- As discussed already in this school, the primary advantage of tungsten lies in its good resistance to sputtering (high mass) and high melting temperature (3695 K). However, both of these benefits argue in direct opposition to good irradiation resistance:



body center cubic structure

3695K melting point

- high mass means higher fraction of defects formed directly within cascade (limiting metallurgical tools to mitigate irradiation effects)

- the application temperature (<1000°C) is < 0.4 Tm puts the material within the low temperature embrittlement regime

• Currently, the manufacturing technology for tungsten and tungsten alloys has struggled to produced "structural" tungsten

Manufacturing of Present Day Tungsten



Texture of Microstructure Highly Dependent on Process

RODS





Properties Can be Highly Non-Uniform

WL10 Rod, Ø7 mm



Microstructure

WL10 Rod, Ø7 mm



• process ductility has been gained by using additions of materials such as La_2O_3 , which resides at grain boundaries.

W Rod, Ø7 mm

Fracture Characteristics of W Rod Materials Ductile W 12 = 12ductile W1Re1La 11 fracture W0.005K 10 -WL10 WL10opt 9 Charpy Energy, J TZM 8 6 4162 5 3 2 brittle **Brittle** fracture 0 -200 300 400 500 600 700 800 900 1000 1100 100 0 Test Temperature, °C

- Rod : Transition from brittle to ductile behavior for powder metallurgy processed materials, in the non-irradiated condition, is above room temperature. Upon irradiation, the transition temperature would further increase.
- Plate : Same situation, though properties are worse.



• Irradiation effects data on tungsten is rather limited, and typically on powder metallurgy samples. Results indicate severe embrittlement at low temperature.

Tungsten Under Irradiation



• Upon irradiation, the transition temperature for which tungsten goes from brittle to ductile behavior increased to well above room temperature.

Importance of Brittle to Ductile Transition Temperature The RMS Titanic

	Titanic hull plate	A36 modern structural steel	
Mn:S Ratio	7:1	15:1 (typical)	

Composition

	Titanic hull plate	A36 modern structural steel
С	0.21	0.20
Mn	0.47	0.55
Р	0.045	0.012
s	0.069	0.01 to 0.04
Si	0.017	0.007
Cu	0.024	0.01
0	0.013	-
Ν	0.0035	0.0032
Mn:S Ratio	7:1	15:1 (typical)

K. Felkins, H. P. Leighly, and A. Jankovic. JOM, 50(1), 1998, 12-18

Importance of Brittle to Ductile Transition Temperature <u>Fracture surface of Charpy</u> <u>specimens from Titanic plate</u>

Longitudinal direction





At 120 °C, ductile fracture

At -32 °C, brittle fracture 20

Effect of temperature on toughness



H. P. Leighly, B. L. Bramfitt, and S. J. Lawrence. Practical Failure Analysis, 1(2), 2001

Tungsten Under Irradiation



• Present forms of tungsten undergo significant embrittlement following low dose irradiation. It currently appears that for end of life ITER and higher dose fusion reactors, tungsten alloys will be fully brittle.

Biased Summary Comment on Tungsten

• Tungsten is potentially a very high performance material. However, its current structural applications (any application) are very limited due to its extreme difficulty in fabrication and limited ductility.

• The poor ductility of tungsten is an issue of both the nature of tungsten crystal, and the nature of the processing used (which controls the grain boundary.) Improvements are possible, and the current understanding of how alloying may improve tungsten ductility, is not well understood.

• Irradiation will make the ductility situation even worse. What is currently needed is a combination of fundamental studies on the irradiation effects

- are in-cascade formed defects a fatal issue for tungsten?
- can be use solid solution alloying to enhance ductility?
- are the current nano-composited tungsten alloys going to be effective

Beryllium as an Engineering Material

• Be has only two operating slip planes {0001} & {1010} significantly limiting the ductility of the metal.

T < 200°C, no {1010} slip, Ductility ~5% 200 < T < 500, {1010} & {0001}, Ductility to 50% T > 500°C, grain boundary failure, Ductility < 20%

- For this reason key engineering properties such as fracture toughness, ultimate strength, and plastic elongation are poor.
- For the non-irradiated case, and especially in the neutron irradiated case, fabrication and design must consider working with a "brittle" material.



Hexagonal Close Packed Crystal

 T_{melt} =1560°C

SP-100 Design Utilized Fairly Complex Beryllium Shapes with Structural and Non-Structural Functions






Effect of Neutron Irradiation on Be Metal



Troev Res. Let Phys 2008

Effect of Neutron Irradiation on Be Metal

The primary effects of neutron irradiation on metallic Be: swelling and embrittlement



Effect of Neutron Irradiation on Be Metal

The primary effects of neutron irradiation on metallic Be: swelling and embrittlement



Defining an Operating Window for Structural Use of Be

- Operating windows are typically defined using following properties:
- Creep
- Fracture toughness
- Fatigue
- Ductility limits (plasticity, elongation)
- Environmental effects
- There are no creep data on modern-day Be, though Larson Miller data exist to estimate the rupture lifetime.
- Design Space Assumptions:
 - Upper stress using extrapolated Larson Miller data.
 - Assume lower bound set by required elongation of 1% total.



Example Fracture Behavior of Beryllium : Fracture Surface

- At elevated temperature, neutron irradiation causes He bubbles to form and embrittle grain boundaries.
- Formation of bubbles and their catastropic effect on mechanical properties have been slightly improved by material purification, but significant embrittlement is unavoidable.
- Example of Improvement:

1960's vintage : 1x10²⁴ n/m² E>0.1 MeV total elongation < 0.2%

1990's vintage : 1x10²⁴ n/m² E>0.1 MeV total elongation few %



000043 5.0 kV X15.0K 2.00 mm







Biased Summary Comments on Beryllium

• Beryllium has been well studied and its use as a plasma facing material requires very high purity materials. Alloying of beryllium will certainly improve mechanical properties, but the consequences on the plasma will likely be unacceptable.

• Improvement in recent years have been largely in the improvement in the grain boundary integrity (removal of oxygen.) Further, incremental improvement is possible, but low temperature embrittlement and helium swelling due to neutron irradiation is unavoidable.

The Graphite Crystal

- The graphite crystal is an interpenetrating hexagonal "benzene" ring structure.
- Very weak bonding between planes, strong bonding in planes.
- Extraordinary in-plane properties, drastically different out-of-plane.

	In-plane	Out-plane
Thermal Conductivity W/m-K)	>2200 W	20
Thermal Expansion	0.5	6.5
Strength (MPa)	>1000	<1
Elastic Modulus(GPa)	20	<1



Irradiated Graphite Crystal

- Interstitials mobile > 70 K, move within the basal plane.
- Vacancies mobile > 1000 K, move freely between the basal plane.
- Interstitial-vacancy recombination barrier < 1 eV (<400K).







Simple Defect Recombination in Graphite : Low Temperature

 As irradiation temperature exceeds activation energy for recombination, vacancy concentration and stored energy is reduced



Simple Defect Recombination in Graphite : Low Temperature

Windscale Air cooled, Graphite Moderated **Operating Temperature ~ 250°C** Core burned to > 1300°C for five days





Dimensional Change in Graphite Crystal High Temperature



 Above ~1000 K both vacancy and interstitials are mobile.

• Dimensional change occurs to high dose <u>conserving volume</u>.

• Higher crystal perfection material suffers less dimensional change due to reduced pinning centers for migrating defects.

Pyrolytic Graphite : Property Changes Under Irradiation



• Both strength and elastic modulus increases pryolytic graphite due to defect pinning.

Nuclear Graphite

H-451 Extruded Nuclear Grade Graphite



IG-11 Isomolded Nuclear Grade Graphite







Effect of Temperature and Swelling of Nuclear Graphite



- initial <c> swelling accommodated by closure of intrinsic porosity.
- once porosity filled swelling can begin.
- less initial porosity for higher initial temperature (closure of intrinsic porosity.)



Effect of Irradiation on Strength of Nuclear Graphite



Effect of Irradiation on Strength of Nuclear Graphite



Effect of Irradiation on Elastic Modulus of Nuclear Graphite

Irradiation-Degraded Thermal Conductivity - Graphite -



- Thermal conductivity in graphite is dominated by phonon transport.
- Vacancy complexes formed during irradiation are extremely effective at scattering phonons and degrading thermal conductivity.



Comparison of Thermal Conductivity Degradation

Thermal Conductivity of Irradiated Ceramics



Thermal Defect Resistance





Thermal defect resistance

Thermal defect resistance due to low vacancy production

The main motivation for using thermal defect resistance is that radiation-induced defects, such as vacancies and clusters, have resistances proportional (or square root dependent) to their concentration and are additive. This gives an easy way to compare stability of ceramics under irradiation.

Graphite Perfection and Thermal Defect Resistance



• Larger, more perfect crystallites accumulate phonon scattering defects at a lower rate in graphite (lower vacancy production.)

---> higher initial conductivity will always have higher irradiated conductivity

Yield Strength of Various Structural Materials



Carbon Fiber Composites

• Composites as being defined here are technically "continuous fiber reinforced composites," the two most mature of which are Carbon(graphite) Fiber Composites (CFC's) and Silicon Carbide fiber composites (SiC/SiC.)

• Of the two, the CFC is the more mature system, though they are similar in terms of processing status and cost.





Reinforced Fired Adobe Composite

Inca city ~ 1500 AD

Present Day





Fort Paramonga Chimu civilization ~1300 AD



Puye Cliff Dwelling Anasaze Indians 1100-1580 AD





Short History of Materials



Fabrication of C/C Composites

Carbon Fiber:

• PAN (polyacrylonitrile) based carbon fiber

- Commercial use for general purpose.
- Varieties: high strength, high modulus, long elongation, ...

Pitch based carbon fiber

- High performance carbon fiber: Anisotropic, high graphitization.
 - Tensile strength: 2.3~4.0GPa, Tensile modulus: 400~900GPa
- General purpose (low cost) carbon fiber: Isotropic microstructure. *Tensile strength: 0.6~1.0GPa, Tensile modulus: 30~60GPa*

Carbon Matrix:

- Chemical vapor deposition (CVD)
- Impregnation and pyrolysis using resin or pitch.

Environmental Barrier Coating: Concern about high reactivity to oxidative products.

- •Boron based glasses (<1000°C)
- Silicon carbide (<1500°C)




Divertor Mock-Ups Using C/C Composites



Full-scale vertical target armored mock-up uses a pure Cu clad DS-Cu tube armored with saddle-block C/C and CVD-W armors. (Hitachi Ltd., Japan)

> Pure Cu clad DS-Cu tube armored with C/C monoblocks. (Kawasaki Heavy Industries, Japan)



Candidate Nuclear Graphite Compared to CFC



	Virgin	500°C	800°C
		$6 \text{ x10}^{25} \text{ n/m}^2$	$7.7 \text{ x} 10^{25} \text{n/m}^2$
Strength	113±20	107 ±7	98 ±11
MPa		(-5%)	(-13%)
Length	-	0.1 %	1.11 %
Change			



	Virgin	500°C	800°C
		$6 \times 10^{25} \text{ n/m}^2$	$7.7 \ge 10^{25} \text{n/m}^2$
Strength	176	286 ±25	241±22
MPa	±20	(+63%)	(+37%)
Length	_	- 1.5 %	-3.6 %
Change			





Dimensional Change in 1-D Composite



Dimensional Change in 3-DComposite



Dimensional Change in FMI-222 Composite



CFC High Dose, High Temperature Irradiation



• Recent, high-temperature studies have shown that the dimensional change in high-quality graphite fiber composite is far more serious than previously thought.

Biased Summary Comments on Graphite and CFC's

- Graphite, like beryllium (and perhaps tungsten) suffers from "crystaldriven irradiation-induced changes. The effects can be somewhat mitigated, but strict lifetimes exist.
- CFC irradiation damage follows the same principals as for graphite.
 - Swelling perpendicular to basal planes, shrinkage within planes
 - Increase in strength and modulus up to composite lifetime.
- The lifetime of the composite will depend sensitively on the irradiation temperature and dose. It appears that, due to the inherent perfection of graphite fibers, the lifetime of the composite may be lower than that of nuclear graphite, especially at high temperatures.
- The CFC materials studied to date have been selected based on high intrinsic perfection (thermal conductivity.) This selection may lead to lower lifetime. --> we may do much better with poorer materials...

Questions ???

CFC's Under Irradiation : Tritium Retention

• T-3 attaches to basal plane edges and highly defected structure. More perfect material and/or high temperature allows less retention.



Operating Window





Effect of Thermo-mechanical Treatment

- Properties of CuCrZr strongly depend on thermo-mechanical treatment
- Optimal heat treatment produces a high number density of fine precipitates, giving rise to high strength



ITER reference heat treatment, SAA: solution anneal at 980-1000°C/0.5-1h, WQ, aged at 460-500°C/2-4 h Heat treatment simulated the manufacturing cycle for large components, **SCA**: HIPpped, SA 980°C/ 0.5 h, slow cooled 50-80° C/min, aged 560 °C/2 h

Meimei Li, M.A. Sokolov, and S.J. Zinkle, (2009).

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