## **Russian Research Center**" Kurchatov Institute"



Investigations of radiation resistance of fission and fusion structural materials in RRC KI using charged particle accelerators

**Alexander Ryazanov** 

**Experience of RRC KI in the Investigations of Materials for Atomic and Fusion Reactors** 

- **Materials for Fusion Reactors:** 
  - C-C, SiC, Al2O3, MgO, ZrO2,
- Metallic Materials for Atomic Reactors:
  - Austenitic Steels
- Atomic Reactors RBMK:

 nuclear graphite, pyrolytic graphite, Zr-Nb
 Basic Research and Theoretical Modeling: Radiation Damage Formation, Defect Cluster Growth, Radiation Swelling, Radiation Creep, Helium Ebrittlement

# Cyclotron of RRC "Kurchatov Institute"





#### **Accelerators of Charge Particles of Russian Research Center "Kurchatov Institute"**

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Cyclotron of RRC KI:
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protons with energy < 35 MeV, current J < 30 mkA
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helium ions He^4 with energy < 60 MeV,
current J < 20 mkA
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ions O<sup>16</sup> with energy < 120 MeV , current J < 5 mkA

ions C<sup>12</sup> with energy < 80 MeV, current J < 5 mkA

# **Radiation Resistance of Graphite Materials**

# Graphite materials in Tokamaks and Fusion Reactor:

#### Limiter and/or protection plate for the inner wall Exposition to:

- Vacuum
- Intense head load
- Edge plasma
- Fast neutrons

#### **Important behavior:**

- Thermal shock resistance
- Thermo-mechanical properties
- Accumulation of Hydrogen and Helium in fusion reactor
- Accumulation of Radiation Damage
- Sputtering and particle emission

# **Graphite Materials for Atomic Reactors :**

Reflectors and stopping system of neutrons

# **Exposition to:**

- Temperature: T = 300-750 C
- Thermal and Fast neutrons

# Important behavior:

- Radiation swelling
- Degradation of thermo-mechanical properties
- Cracking and fracture



# Graphite Sheave after Irradiation in Atomic Station



### **Graphite Elementary Cell**



#### Cell Size Change of Irradiated Pyrographite in Dependence on Irradiation Dose and Temperature



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## **Radiation Shrinkage and Swelling of Graphite**



## **Radiation Damage in Graphite**





## **Experimental Measurement of Radiation Swelling**



#### $\Delta V/V (\Phi_{av}) \cong \Delta Z/d$

 $\Phi_{av}$  – Averaged dpa profile,

- $\Delta Z$  Height of step between irradiated and no irradiated area,
- **d** Penetration depth of irradiated sample.

## **Dose dependence of radiation swelling in SiC**



# Scheme of Irradiation of Graphite Samples by Carbon Ions



#### **Picture of Irradiated and Unirradiated Sample Area, Measurement of Radiation Swelling** Unirradiated **Irradiated area** area 12/01 160 ID# VERT. 200um 0.nm R 163.9um 163.9um 120 Avg 94.95um TIŘ 178.7um Ra 72.52um HORIZ 2000um 80 0.00um R 2000.um 2000.um Angle N/A SCAN MENU 3 40 UM s/UM 10000 .2 2000 .2 1 400 1 5 80 5 25 19 SCAN t=40 s 1500 DIR. -> 500 1000 UM STYLUS 13mg DATA OUT OF RANGE. 0 2000um LEVEL

# **Dose Dependence of Radiation Swelling Pyrolytic graphite at Ec = 3 MeV, T=450 C**



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oa = 1⁄0E21 /h/cm2)

# **Dose Dependence of Radiation Swelling Reactor Graphite at Ec = 3 MeV, T=450 C**



#### **Graphite - two diamenthial anisotropic diffusivity**

Current of point defects on dislocation loop:



graphite

$$J_L = -2\pi b Z_L D_m C_m$$

Current of point defects on spherical void:

$$J_V = -4\pi R_V Z_V D_m C_m$$

Current of point defects on disk void:

$$J_{S} = -2\pi dZ_{S} D_{m} C_{m}$$

Radiation swelling of graphite:

$$S = 2,72C_i - 0,08C_v + 1,242N_LV_L + \Delta \rho_V$$

# Dose dependence of void and loop radius



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# **Dose dependence of swelling**



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# Modeling of dislocation loops in graphite





#### Distribution of displacement field Uz near dislocation loop in graphite as a function of distance from loop



# Bragg scattering in neutron irradiated graphite

#### **Experimental Results**

#### **Theoretical calculations**



# Bragg scattering in C-ion irradiated graphite with energy 20 MeV



# **Radiation Resistance of Ceramic Materials**

#### Dose rate dependence of Ion-induced swelling in CVD-SiC



The displacement damage rates were  $1 \times 10^{-4}$  and  $1 \times 10^{-3}$  dpa/s at 333K-873K and  $1 \times 10^{-5}$  dpa/s at 333K with single-beam irradiation. The error bars represent the 96% confidence limits for the Gaussian distribution. (A. Kohyama)

# Effect of Helium on Radiation Swelling of SiC



Profiles of displacement damage and deposited Ni in irradiated monolithic SiC. Calculated by TRIM-92 assuming Ed=35eV,  $\rho$ =3.21g/cm<sup>3</sup>.



Depth-profiles of atomic displacement damage, deposited He and Ni ions in dualbeam irradiated randomly oriented micro-crystalline SiC calculated by TRIM-92 assuming target mass density of 3.21g/cm<sup>3</sup> and average displacement threshold energy of 35eV.

#### TEM images and SAD patterns for singlebeam (A,B), dual-beam (C,D) and unirradiated (E,F) regions of Hi-Nicalon® Type-S/C/SiC composite

Single-beam 10 dpa 873 K 1x10<sup>-3</sup> dpa/s

Dual-beam 10 dpa 873 K 1x10<sup>-3</sup> dpa/s 60appm-He/dpa

Unirradiated Dark field images from SiC <111> diffraction rings.



#### Dual ion beam irradiation-induced swelling in CVD-SiC



## Theoretical Calculations of effect of Helium on Radiation Swelling in SiC at RRC KI



# He accumulation and irradiation-induced swelling in dual- and single-ion irradiated CVD-SiC at 873K


## **Difference between metals and dielectrics**

### Metals:

- Point defects are neutral
- Electric field does not exist in the matrix

### **Dielectrics (Ceramic Materials):**

- Point defects can have effective charge
- Electric field exists in the matrix under the influence of an applied electric field
- Driving force due to an electric field can have a strong effect on diffusivity of charged point defects





#### Modeling of Dislocation Loops in Ceramics Materials (SiC)



**System of Equations** 

$$D_{m}\Delta C_{m} + \frac{qv_{m}}{kT} D_{m}\nabla(C_{m}\nabla\phi) = 0$$
$$\Delta\phi = -\frac{4\pi}{\varepsilon\omega} \left(\sum_{m} qv_{m}C_{m} + \rho\right)$$

#### **Boundary Conditions**



#### Main parameter values used for numerical calculations of radiation swelling in SiC

$G_1 = G_{Si}$	Point defect generation rate of Si atoms	3.10 <sup>-3</sup> dpa/s
$G_2 = G_C$	Point defect generation rate of C atoms	1.10 <sup>-3</sup> <b>dpa/s</b>
E <sup>Si</sup> mV	Silicon vacancy migration energy	2.3 eV
$E_{mV}^{C}$	Carbon vacancy migration energy	2.0 eV
$E_{ml}^{Si}$	Silicon interstitial migration energy	0.4 eV
$E_{ml}^{C}$	Carbon interstitial migration energy	0.3 eV
E <sup>si</sup> Fv	Silicon vacancy formation energy	2.5 eV
E <sup>c</sup> <sub>FV</sub>	Carbon vacancy formation energy	2.4 eV
$ ho_{ m D}$	Network dislocation density	10 <sup>10</sup> cm <sup>-2</sup>
$\boldsymbol{e}_{v_1} = \boldsymbol{e}_{v_2}$	Vacancy dilatation	-0.1
а	Lattice parameter	$5.14 \times 10^{-8}$ cm

$$D_{VK} = D_{VK}^{O} \exp(-E_{mV}^{K}/T), \text{ (where } D_{V1}^{O} = D_{V2}^{O} = 10^{-2} \text{ cm}^{-2}\text{)},$$
  

$$N_{L} = N_{L}^{O} [\exp(E_{m1}^{1}/T) + \exp(E_{m1}^{2}/T)]^{1/2}, \text{ (where } N_{L}^{O} = 3.10^{12} \text{ cm}^{-3}\text{)}.$$

## Theoretical calculations for time dependence of dislocation loop growth at different irradiation temperatures in SiC





# The comparison of experimental and theoretical temperature dependencies of radiation swelling in SiC.



### INSTABILITY OF INTERSTITIAL CLUSTERS UNDER ION AND ELECTRON IRRADIATIONS IN CERAMIC MATERIAL

#### **Experimental**

- Specimens: 13mol% Y2O3-ZrO2 single crystal (Earth Jewelry Co.)
  - surface orientation: (111)
- □ Irradiation:

- ions: 100 keV He+ at 870 K, up to 1x1020 ions/m2
  - 4 keV Ar+ at 300 K
  - 300 keV O+ at 470-1070 K, up to 5x1019 ions/m2
- electrons: 1000 keV at 470-1070 K, up to 1.4x1027 e/m2
- electron irradiation subsequent to ion irradiation:
  - 100-1000 keV electrons at 370-520 K

#### **Observations:**

- in situ and ex-situ TEM
  - HVEM (JEM-1000, HVEM lab., Kyushu University)
  - TEM (JEM-2000EX, HVEM lab., Kyushu University)
  - TEM-accelerator facility (JEM-4000FX, TIARA, JAERI-Takasaki)

### **Defect clusters in yttrium-stabilized zirconia**

-300 keV O+ions: 5.1x1017 ions/m2 at 470 K -200 keV electrons at 370 K



### **Instability of Interstitial Clusters**



# Characteristic features of the extended defects in yttrium stabilized zirconia

- irradiation condition: under 100-1000 keV electron irradiation subsequent to ion irradiation (100 keV He<sup>+</sup>, 300 keV O<sup>+</sup>, 4keV Ar<sup>+</sup>)
- strong strain and stress fields
- ♦ very high growth rate ≈ 1-3nm/sec
- preferential formation around a focused electron beam
- preferential formation at thick regions
- critical radius: 1.2 μm
  - sudden conversion to the dislocation network
  - repeat nucleation, growth and conversion to dislocation structure on dislocation lines

## **Cross section for displacement in ZrO2**

### under electron irradiation



### Shear stress component induced by charged dislocation loop



 $\sigma^{\rm tot}$ 

## Total normal stress component induced by charged dislocation loop



# Radiation Resistance of Zr-Alloys

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#### Generation Rate of Point Defects under Irradiation of Zr alloy by 15 MeV Helium Ions at Irradiation Dose 10E17cm2



#### Microstructure of Irradiated and unirradiated Zr alloys: 9110 and 9635 (Distribution of β– Nb phases, Laves phases Zr(Nb,Fe)2 and dislocations c-type)

#### **Э110** - β-Nb



#### **3635 Laves phase Zr(Nb,Fe)2**





Microstructure of neutron irradiated Zr russian alloys: Э635 и Э110 up to doses F=0,5x1026n/m2 (2 dpa): distribution of precipitates and ordered dislocation loops (a-type).





Microstructure of Zr alloy 3635 irradiated on cyclotron by protons with energy 4 MeV at T=350oC dose of irradiation 1 dpa (2x10E17p/cm2))



Found optimal irradiation regime allowing to irradiate Zr alloys at T= 300-350 °C up to  $100 \mu m$  deep to doses 10 dpa.

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# Studies of proton beam irradiation on graphite collimator materials for LHC

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Task: Proton Irradiation Damage Assessment of LHC Graphite Collimator Materials

 <u>Main aim of studies</u> – to measure the effect of proton irradiation on physical-mechanical material properties: thermal conductivity, thermal expansion, mechanical properties, electrical resistivity, microstructure change

#### **Objective:**

 Determine the effect of PKA proton energy spectrum near 7 TeV proton beam on physical mechanical properties of graphite collimator material for LHC – irradiation of graphite by protons with the 35 MeV energy at different doses and theoretical modeling of main physical phenomena of radiation effects on materials

#### Neutron energy spectrum per one 7 TeV proton in graphite on the several penetration depths of proton.



# Investigated Graphite Collimator Materials for LHC

 C-C Composite Graphite Material AC-150
 Three orientations:

# **Measured values**

- d density
- λ thermal conductivity coefficient (at T < 700°C)</li>
- ρ electrical resistivity (at T < 700°C)</li>
- α thermal expansion coefficient (at T < 700°C)</li>
- σ compression ultimate tensile stress
- Ed dynamic elastic module
- Es static elastic module
- a, c lattice constants (X-ray method)

#### Scheme of experimental tests of C-C samples on cyclotron



# Target device for proton irradiation of graphite samples on RRC KI cyclotron



- **1-Corps of target device**
- 2-Graphite diaphragm
- **3-Irradiated samples**
- **4-Foil window**
- **5-Holder of samples**
- **6-Main holders**
- **7-Thermocouples**

# Preparation of three orientations of graphite samples for experimental tests



## **Results of physical-mechanical tests (Nº10)**

.№ sample.	d, g/cm3	E, GPa	р, 10 <sup>-6</sup> Ом.м	α, 10 <sup>-6</sup> , 1/Κ
10R(TA-1)	1.66	3.2	27.8	10.31
10R(TA-2)	1.66	2.96	30	10.31
10R(TA-3)	1.66	3.2	29.1	10.31
Average	1.66	3.12	28.9666667	10.31
Magnification	0	0.13856406	1.106044	
10T(AR-1)	1.65	9.3	5.6	0.25
10T(AR-2)	1.64	9.1	5.1	0.25
10T(AR-3)	1.56	9.3	5.7	0.25
Average	1.61666667	9.23333333	5.46666667	0.25
Magnification	0.04932883	0.11547005	0.32145503	
10A(RT-1)	1.54	8.4	5.5	0.167
10A(RT-2)	1.64	8.4	5.7	0.167
10A(RT-3)	1.63	9.16	5.6	0.167
Average	1.603333333	8.653333333	5.6	0.167
Magnification	0.055075705	0.438786205	0.1	)(

### Temperature dependence of electrical resistance for three orientations of graphite samples



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# conductivity for three orientations of graphite



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# Temperature dependence of thermal expansion coefficient of AC-150.



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# Generation rate of point radiation defects under irradiation of graphite by 30 MeV protons at Dose 1.10E17cm2



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# Results of physical-mechanical property changes of graphites after proton irradiation

. <u>№</u> sam pl.	E <sub>0</sub> /E GP a	ΔΕ/E, %	ρ <sub>исх</sub> / ρ <sub>обл</sub> , 10 <sup>-6</sup> Ом.м	Δρ/ρ, %	λ <sub>исх</sub> /λ <sub>обл</sub> Вт/мК	Δλ/λ, %	σ <sub>исх</sub> /σ <sub>обл</sub> МПа	Δσ/σ , %			
First Irradiation ( 0002dpa)											
4R(TA-1)	3,5/4,0	14	28,8/29,65	3	50/48	6	61/65,3	7			
4T(AR-1)	8,1/8,86	9	5,65/8	42	215/150	30	57/59,9	5			
4A(RT-1)	9,2/9,6	4	6,2/9,4	52	215/130	39	60/61,2	2			
Second Irradiation ( 00.2dpa)											
4R(TA-2)	4,8/6,9	42,7	29,9/62,5	109	51/25	51	61/73,8	21			
4T(AR-2)	9,3/10	7,5	5,7/17	198	215/80	62	57/60,4	6			
4A(RT-2)	8,8/9,3	5,7	5,75/12	109	215/105	51	60/62,4	4			
Third Irradiation (02dpa)											
4R(TA-3)	3,7/5,5	48,8	27.6/108	290	51/15	-70	61/74,4	22			
4T(AR-3)	8,3/9,1	9,6	5.56/26,8	370	215/55	-74	57/59,9	5			
1 A (DT 2)	0 1/10 2	9.5	5 5/26 5	201	245/56	72	60/62 A	4			

# Dose dependence of radiation swelling for three orientations of graphite.



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# Dose dependence of elastic Yung module for three orientations of graphite.



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D,dpa

# Dose dependence of thermal expansion coefficient for three orientations of graphite.



D, dpa



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# Temperature dependence of thermal expansion coefficient of AC 150.



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#### Generation Rate of Point Defects under Irradiation of Graphite by 5 MeV Carbon Ions at Irradiation Dose 5.10E17cm2



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## Scheme of Irradiation of Graphite Samples



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Measurements of Radiation Induced Deformation in Graphite Composite Material REC Irradiated by Carbon Ions with the Energy 5 MeV at Irradiation Dose: 3x10 E17 p/см 2



Measurements of Radiation Induced Deformation in Graphite Composite Material AC Irradiated by Carbon Ions with the Energy 5 MeV at Irradiation Dose: 1x10 E17 p/см 2



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#### Measurements of Radiation Induced Deformation in Graphite Composite Material R4SSO Irradiated by Carbon Ions with the Energy 5 MeV at Irradiation Dose: 3x10 E17 p/см 2



Measurements of Radiation Induced Deformation in Pyro -Graphite Material Irradiated by Carbon Ions with the Energy 5 MeV at Irradiation Dose: 1x10 E17 р/см 2



#### Dose Dependence of Radiation Swelling of Graphite Collimator Materials of LHC



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### Dose Dependence of Radiation Swelling of Pyro-graphite Material



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Future tests for of physical – mechanical properties changes on proton irradiated Graphite Collimator Materials for LHC

The following measurements shall be performed in all three directions of the irradiated C-C graphite materials (where applicable):

- Thermal conductivity (to  $\pm 5 \text{ W/m/K}$ ).
- Electrical resistivity (to  $\pm 1 \text{ m}\Omega \text{ m}$ ).
- Thermal expansion coefficient (to  $\pm 10^{-6} / {}^{\circ}C$ ).
- Mechanical strength (elastic modulus, deformation to rupture, ultimate tensile and compression stress, yield stress).
- Microstructure change with scale (visual analysis).
- Radiation erosion.
- Density and specific heat.
- Dimensions.

Make analysis with TEM, STEM, radiography and mechanical and electrical test equipment on irradiated and non-irradiated samples.

• Make theoretical analysis (dpa levels per sample and help in predictions for the LHC irradiation conditions).



Theoretical modeling of shock wave formation under 450 GeV and 7 TeV proton beams in collimator materials for LHC

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#### Conclusion

- The results presented here allow to clarify main physical mechanisms of radiation resistance of fission and fusion structural materials using charged particle irradiation on accelerators.
- Using accelerators of charged particles allow to investigate the following physical phenomena in irradiated materials for fission and fusion reactors:
  - Radiation hardening
  - Radiation swelling
  - Irradiation creep
  - Helium and hydrogen embrittlement
  - Fracture processes under irradiation
  - at high irradiation doses and different temperatures.
- The changes of physical-mechanical properties for the following fission and fusion structural materials can be analyzised:
  - Graphite and C-C composites
  - Ceramic materials: SiC, MgO, ZrO2, Al2O3
  - Zr alloys
  - Ferritic / martensitic and austenitic steels
  - ODS materials
  - V alloys
- Using accelerators can help to make chose more radiation resistance materials for fission and fusion reactors during short irradiation time comparing with atomic reactors