In situ irradiation testing of nuclear ceramics and oxides with heavy ions of fission fragments energy

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• Introduction
• High energy heavy ion irradiation facility in FLNR JINR
• Study of structural effects of dense ionization in nuclear ceramics and oxides with heavy ions of fission fragment energy
• Real time examination of mechanical stress in Al$_2$O$_3$ under swift heavy ion irradiation
• Residual stress depth profiles in oxide materials irradiated with high energy heavy ions
• Outlook
Examination of the dense ionization effect in ceramics and oxides with heavy ions of fission fragments energy

The overall intention of this work is to yield sufficient basic data to determine and compare the radiation tolerance of several ceramics and single crystals (MgAl$_2$O$_4$, MgO, Al$_2$O$_3$, ZrO$_2$, SiC, ZrC, AlN, Si$_3$N$_4$) considered as candidates for inert matrix fuel hosts.

Our central objectives are:
- to study the temperature dependence of swift heavy ion-induced phase transformations and dense ionization effect on pre-existing defect structure in irradiating materials
- to elucidate the correlation between surface and material bulk radiation damage induced by heavy ions with energies above 1 MeV/amu
- real time examination of stress accumulation in ceramic materials under swift heavy ion bombardment
Ion tracks in spinel irradiated with 430 MeV Kr ions to a fluence of \(1.1 \times 10^{12}\) cm\(^{-2}\) at room temperature. The average TEM track diameter is \(\sim 2\) nm.

Threshold ionizing radiation levels for track formation in ceramics

<table>
<thead>
<tr>
<th>Material</th>
<th>$S_e$, keV/nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>MgAl$_2$O$_4$</td>
<td>8</td>
</tr>
<tr>
<td>Si$_3$N$_4$</td>
<td>15</td>
</tr>
<tr>
<td>Al$_2$O$_3$</td>
<td>&gt; 41</td>
</tr>
<tr>
<td>AlN</td>
<td>&gt; 34</td>
</tr>
<tr>
<td>SiC</td>
<td>&gt; 34</td>
</tr>
</tbody>
</table>

High-resolution lattice image of Si$_3$N$_4$ irradiated with 710 MeV Bi ions (plan-view specimen)

High-resolution lattice image of $\alpha$-Al$_2$O$_3$ irradiated with 710 MeV Bi ions a fluence of $7 \times 10^{12}$ cm$^{-2}$

The average TEM track diameter is $\sim$3 to 4 nm.

TEM micrograph of $\alpha$-A$_2$O$_3$ target irradiated at $S_e=41$ keV/nm to a fluence of $7 \times 10^{12}$ cm$^{-2}$ (S. J. Zinkle, ORNL)

The presence of numerous subgrains suggests that considerable internal stresses were induced by the Bi ion irradiation
Examination of the dense ionization effect in ceramics and oxides with heavy ions of fission fragments energy

The mean positron lifetime as a function of dose. The figure shows different stages of point defect accumulation.
Examination of the dense ionization effect in ceramics and oxides with heavy ions of fission fragments energy

3D AFM image of MgAl$_2$O$_4$ surface irradiated with 710 MeV Bi ions. Ion fluence $5 \times 10^{10}$ cm$^{-2}$. 
Mean hillock height versus incident electronic energy deposition

Threshold electronic stopping power value needed for the hillocks production:

- MgO, $S_e \approx 15.8$ keV/nm
- MgAl$_2$O$_4$, $S_e \approx 15.5$ keV/nm
- Al$_2$O$_3$, $S_e \approx 25$ keV/nm
- SiC, $S_e > 34$ keV/nm
Main questions addressed to real time measurements:

- Radiation damage and stress accumulation processes before and after ion track region overlapping
- Variation in the stress state under ion irradiation characterized by specific ionizing energy losses higher and lower than the threshold of radiation damage formation via electronic excitations.
FLNR cyclotron complex

U400M $E = 6 \div 100$ MeV/n

U400 - $E = 0.5 \div 20$ MeV/n

IC-100 - $E \approx 1.2$ MeV/n

U200 - $E = 3 \div 15$ MeV/n
This heavy ion irradiation facility is suitable for irradiating large area (10x60 cm) polymer films just as small metal, semiconductor and ceramic samples in well controlled circumstances.

A homogeneous ion beam distribution has been achieved using horizontal and vertical high-frequency electrostatic or low-frequency electromagnetic scanning systems. Ion beam homogeneity is better than 5%.
$B^{+2}$, $Ne^{+4}$, $Ar^{+7}$, $Fe^{+8}$, $Kr^{+15}$, $I^{23}$, $Xe^{+23}$, $W^{+32}$ ions with energy $\approx 1.2$ MeV/n
SEM data: d=0.2 μm
Experimental set-up for ion-beam-induced luminescence measurements on IC-100 FLNR JINR cyclotron
Basis of the piezospectroscopic effect – the applied stress strains the lattice and alters the energy of transitions between electronic states

\[ \Delta \nu = \Pi_{ij} \times \sigma_{ij} \]

\( \Pi_{ij} \) – piezospectroscopic coefficients

Typical piezospectroscopic probes: Cr\(^{3+}\) in Al\(_2\)O\(_3\)
Eu\(^{3+}\), Nd\(^{3+}\) in silica glasses
Sm\(^{3+}\) in borosilicate glasses
Ion irradiation parameters

<table>
<thead>
<tr>
<th>Ion type and energy, MeV</th>
<th>T, K</th>
<th>Φ, cm(^{-2})s(^{-1})</th>
<th>P, W cm(^{-2})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ar(^{16}), 280</td>
<td>300</td>
<td>3 × 10(^8)</td>
<td>0.013</td>
</tr>
<tr>
<td>Kr(^{26}), 245</td>
<td>80</td>
<td>300(I)</td>
<td>6.3 × 10(^8)</td>
</tr>
<tr>
<td></td>
<td>300 (II)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Bi(^{51}), 710</td>
<td>80</td>
<td>300</td>
<td>1.3 × 10(^8)</td>
</tr>
</tbody>
</table>

\[ \Delta \nu = \Delta \nu(\sigma) + \Delta \nu(T) + \Delta \nu(n_{Cr}) \]

the hydrostatic stress component

\[ \sigma_h = (\sigma_{11} + \sigma_{22} + \sigma_{33})/3 \approx \Delta \nu/7.61 \]

\( \sigma_h \) (GPa), \( \Delta \nu \) (cm\(^{-1}\))
The magnitude of integrated stress in Al$_2$O$_3$ induced by hundred keV- some MeV ion irradiation depends on the ratio of electronic to nuclear stopping power.


The knowledge about of high energy heavy ion-induced stress is of considerable practical value in view of simulation of fission product impact in radiation resistant oxides and ceramics, considered as candidate materials for nuclear waste management.
Dose dependence of the \( R \)-lines spectra under 167 MeV Xe ion irradiation. Ion beam incidence angle is 60\(^\circ\). \( T=80\) K.
The $R$-lines spectra measured during Kr, Xe and Bi tilted ion bombardment as a function of ion fluence. Spectra are normalized on maximum of the $R_1$-line intensity.


670 MeV Bi  $S_e$=41 keV/nm
167 MeV Xe  $S_e$=24.7 keV/nm
107 MeV Kr  $S_e$=16.4 keV/nm

! No stress relaxation occurs if $S_e$ less than threshold value of damage formation via electronic excitation
Dose dependence of the difference between current and first registered $R_1$-line spectrum during 167 MeV Xe ion irradiation
Dose variation of the $R$-lines spectra under 167 MeV Xe and 107 MeV Kr ion irradiation at normal ion beam incidence

The splitting of the $R$-lines directly indicates the presence of differently stressed regions in the irradiating ruby specimen.

Each Cr$^{3+}$ ion acts as an independent strain sensor.

The stresses are compressive in basal plane of the sample and tensile in perpendicular direction.
Laser Confocal Scanning Microscopy (LCSM) Study of Residual Stress Profiles in $\text{Al}_2\text{O}_3$:Cr After Swift Heavy Ion Irradiation

Conventional optical microscope

Confocal microscope

LCSM Solar TII
Photoluminescence R-lines spectra registered in virgin and 710 MeV Bi ion irradiated ruby specimens
Stress tensor components and ionizing energy loss profiles in ruby

The signs of the stress tensor components indicate that stresses are compressive in basal plane of the sample and tensile in perpendicular direction.
Depth-resolved R-lines photoluminescence spectra measured using LCSM technique (postradiation examination)

240 MeV Kr, 300 K, ion fluence $-10^{14}$ cm$^{-2}$

virgin