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Effects of swift heavy ion irradiation and simulation of fission fragment impact

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Element 114 is Named Flerovium and Element 116 is Named Livermorium

IUPAC has officially approved the name flerovium, with symbol Fl, for the element of atomic number 114 and the name livermorium, with symbol Lv, for the element of atomic number 116.

Priority for the discovery of these elements was assigned to the collaboration between the Joint Institute for Nuclear Research (Dubna, Russia) and the Lawrence Livermore National Laboratory (Livermore, California, USA).

The name flerovium will honor the Flerov Laboratory of Nuclear Reactions where superheavy elements are synthesised. Georgiy N. Flerov (1913 – 1990) – was a renowned physicist, author of the discovery of the spontaneous fission of uranium, pioneer in heavy-ion physics, and founder in the Joint Institute for Nuclear Research the Laboratory of Nuclear Reactions (1957).

The name livermorium honors the Lawrence Livermore National Laboratory. A group of researchers of this Laboratory with the heavy element research group of the Flerov Laboratory of Nuclear Reactions took part in the work carried out in Dubna on the synthesis of superheavy elements including element 116.

Dubna: the root – "dub" – oak tree



Swift heavy ions – ions with energies near and above 1 MeV/nucleon

Main sources of high energy ions:

- I. Accelerators
- **II.** Galactic cosmic rays
- **III.** Fission fragments

I. Accelerators

High energy heavy ion accelerator facilities:

GANIL (Caen, France) 1-100 MeV/amu GSI (Darmstadt, Germany) 10-1000 MeV/amu NSC (New Delhi, India) 15 MeV×Z JINR (Dubna, Russia) 1.2-10 MeV/amu CRC (Louvain-la-Neuve, Belgium) 0.6-27.5 MeV/amu ANU (Canberra, Austarlia) 15.5 MeV×Z RIKEN (Tokai, Japan) 7-135 MeV/amu JAERI (Takasaki, Japan) 2.5-90 MeV/amu IRC (Astana, Kazakhstan) 0.42-1.67 MeV/amu BNL (Berkeley, USA) 4.5-55 MeV/amu

Accelerator for applied research – IC100



B⁺², Ne⁺⁴, Ar⁺⁷, Kr⁺¹⁷, Xe⁺²⁶ ions with energy \approx 1.2 MeV/amu Ion fluence range – up to 10¹⁶ cm⁻²



U400M FLNR JINR Cyclotron



Ion Beam Line for SEE Testing at U400M FLNR JINR Cyclotron



lons ¹⁶O ÷ ²⁰⁹Bi, E= 3 ÷ 9 MeV/nucleon, LET = 4 ÷ 100 MeV/(mg/cm²)

DC-60 CYCLOTRON (Astana)



II. Galactic cosmic rays

The composition of galactic cosmic rays includes: 83% protons 13% alpha particles 3% electrons 1% heavy ions (Z>4)



III. Fission fragments



Nuclear Instruments and Methods in Physics Research B35 (1988) 513-517 North-Holland, Amsterdam

Section VI. New techniques and developments

FRAGMENTS FROM FISSION: THE FORGOTTEN IONS

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Mass distribution of fission fragments from Pu²³⁹. Solid line is coincident fragment experiment; dashed line is chemical analysis.

D.C.Brunton and W.B.Thomson. Phys. Rev. 76(1949), 848



Energy spectrum of U²³⁵ fission fragments induced by thermal (a) and 2.5 MeV (b) neutrons. *Stephen S. Friedland. Phys. Rev.* 84(1951), 75



TEM image of tracks from ²³⁵U fission fragments in a synthetic fluor-phlogopite mica. *R.L., P.B. Price, R.M. Walker: "Nuclear Tracks in Solids: Principles and Applications." University of California, Berkeley, 1-605, (1975)* Main peculiarity of swift heavy ion interaction with solids is a high level of ionizing energy losses which may result in formation of specific radiation damage – latent tracks



Aim of presentation – to give a review on structural effects of dense ionization in some nuclear ceramics and oxides under swift heavy ion irradiation simulating fission fragment impact

Materials to be considered – candidate materials for inert matrix fuel hosts (IMF)

Inert matrices - ceramics with a high melting point and with low neutron absorption cross sections to be used as hosts for transmutation of actinides via nuclear reactions

Ceramics and oxides considered as candidates for inert matrix fuel hosts - $MgAl_2O_4$, MgO, Al_2O_3 , ZrO_2 , SiC, ZrC, ZrN, AlN, Si_3N_4

Examples of fuel compositions for LWR: (Pu)O₂ - MgO-ZrO₂ (Pu,Np,Am)O₂, MgO-ZrO₂ (Am,Np,Pu,Zr)N



Phase transformations and accompanying volume changes in swift ion/(fission fragment track region may produce unacceptable stresses in fuel pin assemblies

Al₂O₃: $\rho_{crystalline} / \rho_{amorphous} = 0.03$

Our central objectives are:

- Threshold electron stopping power for radiation damage formation via electronic excitations -?

- single ion track and ion track overlapping irradiation regimes-?

 dense ionization effect on pre-existing defect structure in irradiating materials - ?

- correlation between surface and material bulk radiation damage induced by heavy ions with energies above 1 MeV/amu - ?

- evaluation of mechanical stresses in oxides -?

$MgAl_2O_4$



C. Kinoshita. Microstructural evolution of irradiated ceramics. In: Radiation effects in solids. Eds. K.Sickafus, E.Kotomin and B.Uberaga. Nato Science series. V.232, p.193

spinel is not amorphized by electron, neutron and low-energy ion irradiation at T > 300 K

$MgAl_2O_4$



Ion tracks in spinel irradiated with 430 MeV Kr ions to a fluence of 1.1×10^{12} cm⁻² at room temperature.

The average TEM track diameter is ~2 nm.

S.J. Zinkle, V.A. Skuratov . NIMB. B 141 (1998) 737





Ion tracks in spinel irradiated with 430 MeV Kr ions

Swift heavy ions induce discontinuous nonamorphous tracks

S.J. Zinkle, V.A. Skuratov . NIMB. B 141 (1998) 737



HR lattice images of spinel irradiated with 5 ×10¹¹ 200 MeV Xe ions/cm²

T. Yamamoto et al. / Nucl. Instr. and Meth. in Phys. Res. B 245 (2006) 235–238



The calculated electronic and nuclear stopping power profiles in $MgO \cdot 1.1Al_2O_3$ irradiated with 200 MeV Xe and 350 MeV Au ions

Track core size : 2-3 nm Strained regions size: (around 5 nm)

The size of the disordered regionsXe, 200 MeVAu, 350 MeV12.8±0.9 nm for Al ions10.6±0.7 nm for Al ions9.6±0.4nm for Mg ions8.8±0.5 nm Mg ions

The disordered regions are around 10 nm in diameter

K. Yasuda, T. Yamamoto and S. Matsumura. JOM Journal of the Minerals, Metals and Materials Society 59(2007), N4, 27.





Bright-field cross section view of MgO·1.1Al₂O₃, illustrating continuous ion tracks along incident ions (a), irradiated at 300 K with 350 MeV Au ions to a fluence of 5×10^{11} ions/cm², and HARECXS profiles taken from an unirradiated specimen (b) and from a depth of 2 µm in the irradiated specimen (c) and from a depth of 8 µm (d).

Diameter of the disordered region induced by one 350 MeV Au ion in MgO \cdot 1.1Al₂O₃ as a function of electronic stopping power

K. Yasuda et al. / NIM B 266 (2008) 2834–2841



The volume fraction of disordered damage region of MgO $\cdot 1.1$ Al₂O₃ irradiated with 200 MeV xenon and 350 MeV gold ions against of ion fluence.

K. Yasuda, T. Yamamoto and S. Matsumura. JOM Journal of the Minerals, Metals and Materials Society 59(2007), N4, 27.



Cross-section microstructure of $MgAl_2O_4$ irradiated to a fluence of $1x10^{16}$ 72 MeV lodine ions/cm²

volumetric swelling is ≈35% for the crystalline to amorphous phase transformation

Heavy ions of fission products energy can amorphize spinel. Ion fluence threshold for amorphization is between 1x10¹³ and 5 1x10¹³ cm⁻²

Ion track region area for R=1 nm =10⁻⁷ cm S = 3.14× 10⁻¹⁴ cm² Ion fluence needed to cover area 1 cm² Φ = 1/3.14×10⁻¹⁴ cm² ≈ 3.18×10¹³ cm⁻² R=2 nm, Φ =1/12.56 ×10⁻¹⁴ cm² ≈ 7.9×10¹² cm⁻²

Proc. 540 (1999) 299.

Tungsten deposited



XTEM image of polycrystalline MgAl₂O₄ sample irradiated with 85 MeV I⁺⁷ ions to 1.2×10^{15} cm⁻²

Electronic and nuclear stopping powers, Se, Sn of $MgAl_2O_4$ for incident 85 MeV I⁺⁷ ions as a function of depth. Ion fluence 1.2×10^{15} cm⁻²

T. Aruga et al. / NIM B 197 (2002) 94–100



Step heights formed on the surfaces of MgAl₂O₄ and Al₂O₃ along a border of masked and 85 MeV I⁺⁷ ion irradiated area. Ion fluence 1.2×10^{15} cm⁻²

The swelling due to amorphization is estimated to be around 15–20%

T. Aruga et al. / NIM B 197 (2002) 94–100



XTEM image of polycrystalline MgO sample irradiated with 85 MeV I⁺⁷ ions to 0.28×10^{15} cm⁻² (a) and 1.2×10^{15} cm⁻² (b)

No amorphization occurs for MgO samples at the fluence of 1.2×10^{15} cm⁻²

T. Aruga et al. / NIM B 197 (2002) 94–100

Dense ionization effect on pre-existing defect structure in MgAl₂O₄

MgAl₂O₄ + 2 MeV Al , 950 K, 30-100 dpa Defect structure – interstitial loops ($\emptyset \sim 30 \text{ nm}$, N $\sim 4 \times 10^{15} \text{ cm}^{-3}$), Al precipitates (2÷25 nm, N $\sim 10^{15} \text{ cm}^{-3}$)

MgAl₂O₄ + 3.6 MeV Fe , 950 K, 1-6 dpa Defect structure – interstitial loops ($\emptyset \sim 5 \text{ nm}$, N $\sim 2 \times 10^{17} \text{ cm}^{-3}$)

the loop size had decreased to 4 nm and the loop density had increased to N $\sim 4 \times 10^{17}$ cm⁻³

Swift heavy ion irradiation induces dissolution of dislocation loops which size is comparable with latent track diameter

+ 430 MeV Kr 1.1×10¹² cm⁻²

S.J. Zinkle, V.A. Skuratov . Nucl. Instr. and Meth.. B 141 (1998) 737



Microstructure beyond the end of range of the 2 MeV Al ion preirradiated region in spinel after 430 MeV Kr ion irradiation

S.J. Zinkle, V.A. Skuratov . Nucl. Instr. and Meth.. B 141 (1998) 737

Summary on MgAl₂O₄:

Threshold ionizing radiation levels for track in spinel - $6 \div 8 \text{ keV/nm}$ Multiple overlapping of the ion track regions leads to amorphization of MgAl₂O₄ High energy heavy ion irradiation may induce dissolution of dislocation loops which size is comparable with latent track diameter

Ion	Energy,	Fluence	dF/dx,	Ref.	Swelling,	Track radius,	Accelerator
	MeV	ions, cm ⁻²	keV nm ^{·1}		%	nm	
127 _I	70	1011	16	this work		1.5	¹ TU Munich
127 _I	70	1013	16			Not observed	-
¹²⁷ I	70	5·10 ¹²	16	**	0		*
¹²⁷ I	70	5·10 ¹³	16	- 14	11.4 (7.25)*		*
127I	70	5·10 ¹⁴	16	44	22.2 (13.5)*		it.
¹²⁷ I	70	5·10 ¹⁵	16	44	33.1 (21)		
²⁰⁹ Bi	120	5·10 ¹⁰	23	"		3.4	² GSI
²⁰⁹ Bi	2380	5·10 ¹⁰	34			1.7	GSI
²³⁸ U	2710	5·10 ¹⁰	41	**		1.5	GSI
⁸⁶ Kr	430	1.1 1012	16	[13]		1	³ U-400
¹²⁹ Xe	614	6.10 ¹¹	26	[13]		1.3	U-400

TANDEM of Beschleunigerlaboratorium der LMU und TU München,

² UNILAC of Gesellschaft für Schwerionenforschung, Darmstadt

³U-400 cyclotron of Joint Institute for Nuclear Research, Dubna.

* The swelling values in brackets are previously reported results [12] which were obtained using the ion full range.

T. Wiss et al. Progress in Nuclear Energy. Vol. 38, NO. 3-4, pp. 281-286, 2001

 Al_2O_3



Evolution of the damage cross-section vs electronic stopping power Al_2O_3 . The threshold of damage formation through dense ionization is about **20 keV/nm** **RBS-C** analysis

 α –relative disorder near the sample surface

 $\alpha = \frac{\chi_0 - \chi_v}{1 - \chi_v}$

 χ_0 – dechanneling yield χ_0 - minimum yield

$$\alpha = 1 - \exp(-A_e \Phi)$$

A_e – damage cross-section

B.Canut et al. PRB Vol. 51, No. 18, pp. 12194-12201, 1995



²³⁸U (115 MeV, 2.46×10¹² cm⁻²) + α -Al₂O₃

The major part of F-type defects is created by elastic processes

Evaluation of the F-type defect concentration from optical absorption spectra

 $N = 1.31 \times 10^{16} (A \times W_{1/2})/f$

A -absorbance at peak position $W_{1/2}$ is the full width at the half maximum of the absorption peak (eV), **f** – oscillator strength

B.Canut et al. PRB Vol. 51, No. 18, pp. 12194-12201, 1995



1.157 GeV ⁵⁶Fe (•), 1.755 GeV ¹³⁶Xe(), 2.636 GeV ²³⁸U())

The nuclear energy loss processes determines the production of F-type defects in swift heavy ion irradiated Al₂O₃

Y. Song et al. / NIMB 254 (2007) 268–272

Mean positron lifetime vs. Ion fluence in irradiated sapphire 240 Kr 250 MeV 230 Bi 710 MeV Ο 220 210 200 τ_{mean} (ps) 190 Ο 0 180 Electron baitros 170 160 substrate 150 1E10 1E11 1E12 1E9 1E13 1E14 Fluence (ions cm^{-2}) trapping in trapping in saturated vacancies larger trapping in defects vacancies

The figure shows different stages of point defect accumulation



Zs. Kajcsos: Positron annihilation studies...(2004)





HREM micrograph of sapphire irradiated with 30 MeV ⁶⁰C²⁺ (a) and 10 MeV ⁶⁰C⁺ (b) at normal incidence.





(a) Plane view micrograph of sapphire
irradiated with 10¹⁰ ⁶⁰C²⁺ cm⁻² at 30 MeV. (b)
The histogram shows the repartition of the
track diameters

S.M.M. Ramos et al. / NIM B 143 (1998) 319-332



High-resolution lattice image of α -A₂O₃ irradiated with 710 MeV Bi ions (plan-view specimen). Ion fluence 7x10¹² cm⁻². S_e =41 keV/nm

S. Zinkle, ORNL



Surface topography image of Al₂O₃ irradiated by 1 MeV/u Au ions (5×10⁹ ions/cm²). Right: height versus diameter of hillocks.

N. Khalfaoui et al. NIMB, in press



XTEM micrograph of Al_2O_3 irradiated with 85 MeV I^{7+} ions to 0.28×10^{15} cm⁻²

Nuclear and electronic stopping power depth profiles for 85 MeV I ions incident on Al_2O_3

T. Aruga et al. / NIM B(166-167)(2000),913-919



XTEM micrograph of A₂O₃:Cr single crystal irradiated with 167 MeV Xe ions. Φ t=2.9×10¹³ cm⁻², T=80 K, ion beam incidence angle 30°, R_p≈ 5 µm

V.A. Skuratov et al . NIMB. B 268 (2010) 3023



Fluence dependence of the relative disorder in irradiated sapphire





Evolution of the completely amorphous thickness a versus fluence deduced from RBS-C measurements.

Step height of swelling versus ions fluence for α -Al₂O₃ **H=H₀(1-exp(\sigma_{sw}\Phi)** σ_{sw} the effective swelling cross section

RBS-C analysis evidences the presence of two processes: partially disordered tracks first overlap, and in a second step an amorphous layer is growing linearly with fluence from the sample surface.

A. Kabir et al. / NIM B266(2008)2976–2980



Variation of the lattice strain versus the fluence for α -Al₂O₃ irradiated with 90.3 MeV xenon ions

Swelling leads to build-up of mechanical stresses in the irradiating material

A. Kabir et al. / NIMB 268 (2010) 3195–3198

Swelling of insulators induced by swift heavy ions



Profilometer scans for Pb (4 MeV/u) irradiations of $Y_3Fe_5O_{12}$ at different fluences (ions/cm²).

Step height versus fluence for different crystals irradiated with Pb ions of 4 MeV/u

C. Trautmann et al. / NIMB 191 (2002) 144–148



yttrium stabilized zirconia (YSZ)



Variation of the hydrostatic and biaxial stress components in CSZ irradiated with 940 MeV Pb ions.

Sattonnay et al. J. Appl. Phys. 101, 103516 2007

Crystalline to crystalline phase transformation induced in oxides by swift heavy ions

Yttrium oxide Y₂O₃ : cubic to the monoclinic phase (1-GeV Ta, 0.86-GeV Pb ions)

S.Hemon/ NIMB 146 (1998) 443-448

Pure zirconia ZrO_2 : monoclinic to the tetragonal phase (Se > 13 keV/nm)

A. Benyagoub / NIMB 206 (2003) 132-138



Evolution of the fraction of the tetragonal phase with the ion fluence in the case of the irradiation of pure zirconia with 135-MeV Ni ions.

yttrium stabilized zirconia (YSZ)



Accumulated damage (fd) versus ion fluence in YSZ crystals irradiated with 4MeV Au²⁺ and 940 MeV Pb⁵³⁺ ions

S. Moll et al. / NIM B 266 (2008) 3048–3051



Accumulated damage (*fD*) versus ion fluence for nonamorphizable (a) and amorphizable (b) ceramics irradiated at RT with slow ions. Insets show TEM diffraction patterns recorded at the final fluences on both types of materials.

L. Thome et al. Advances in Materials Science and Engineering Volume 2012 (2012), Article ID 905474, 13 p.



Accumulated damage (fD) versus ion fluence for nonamorphizable (a) and amorphizable (b) ceramics irradiated at RT with swift ions. The inset shows a TEM diffraction pattern recorded at the final fluence on $Gd_2Ti_2O_7$.

> L. Thome et al. Advances in Materials Science and Engineering Volume 2012 (2012), Article ID 905474, 13 p.



Electronic sputtering of nitrides by high-energy ions



The sputtering yields are larger by 30 -2×10³ than those of calculations based on the elastic collision cascades

Sputtering yields at Se = 15 (o) and 5 keV/nm (\bullet) as a function of the band-gap The electronic sputtering yields scale with the electronic stopping power as S_e^n with n = 1.4-4

> N. Matsunami et al. / NIMB 209 (2003) 288-293 N. Matsunami et al. / NIMB 256 (2007) 333–336

Ionizing energy loss threshold values for latent track formation in nitrides and carbides

Vaterial S _{et} , keV/nm		Ref.		
AIN	> 34	S.J. Zinkle, J.W. Jones, V.A. Skuratov. MRS Symposium Proceedings, Vol. 650, MR, Warrendale, PA, 2001, p. R3.19.1. S.J.Zinkle, V.A.Skuratov and D.T.Hoelzer. NIMB 191(2002) 758		
	> 21.6	S. Mansouri et al. // NIMB 266(2008) 2814		
BN	> 17.6	S. Mansouri et al. // NIMB 266(2008) 2814		
SiC	>34	S.J. Zinkle, J.W. Jones, V.A. Skuratov. MRS Symposium Proceedings, Vol. 650, MR, Warrendale, PA, 2001, p. R3.19.1.		
SiC Nano-SiC	> 22 > 17	A. Audren et al. // NIMB 267(2009) 976, NIMB 266 (2008) 2806		
Nano-ZrN	> 49	A. Janse van Vuuren, V. A. Skuratov, V.V. Uglov (in press)		
Si ₃ N ₄ 15		S.J. Zinkle, J.W. Jones, V.A. Skuratov. MRS Symposium Proceedings, Vol. 650, MR, Warrendale, PA, 2001, p. R3.19.1. S.J.Zinkle, V.A.Skuratov and D.T.Hoelzer. NIMB 191(2002) 758		



Mid-range microstructure of Si_3N_4 irradiated with 710 MeV Bi ions (cross-section specimen)



High-resolution lattice image of Si₃N₄ irradiated with 710 MeV Bi ions (plan-view specimen)



S.J.Zinkle, V.A.Skuratov and D.T.Hoelzer. NIMB 191(2002) 758



Silicon carbide

XTEM images of *n*-4*H*-SiC CVD epitaxial layer irradiated with Bi ions. E = 710 MeV, 5 10^{10} cm⁻²

CVD layer thickness – 26 μ m R_p = 29 μ m

end of range area annealing at 500C

end of range area

as irradiated

subsurface

layer

E.V. Kalinina, V.A. Skuratov et al. Semiconductors, v.41 (2007), iss. 4, 396

Dense ionization effect on pre-existing defect structure in SiC



ion-beam-induced epitaxial crystallization (IBIEC effect)

Epitaxial recrystallization induced by the irradiation with 827 MeV Pb ions of SiC samples previously damaged with 700 keV I at the fluence of 9×10¹³ I cm⁻²

A. Benyagoub and A. Audren J. Appl. Phys. 106, 083516 2009

Thank you for your attention!