Nuclear Instruments and Methods in Physics Research B 314 (2013) 195–201

Contents lists available at SciVerse ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Computer simulation of structural modifications induced by highly energetic ions in uranium dioxide

Y. Sasajima [a,b,*], T. Osada [c], N. Ishikawa [d], A. Iwase [e]

[a] Department of Materials Science and Engineering, Faculty of Engineering, Ibaraki University, 4-12-1 Nakanarusawa, Hitachi 316-8511, Japan
[b] Frontier Research Center for Applied Atomic Sciences, Ibaraki University, Shirakata 162-4, Tokai 319-1106, Japan
[c] Graduate School of Science and Engineering, Ibaraki University, 4-12-1 Nakanarusawa, Hitachi 316-8511, Japan
[d] Japan Atomic Energy Agency (JAEA), Shirakata Shirane 2-4, Tokai 319-1195, Japan
[e] Department of Materials Science, Osaka Prefecture University, Gakuen-cho 1-1, Sakai 599-8531, Japan

ARTICLE INFO

Article history:
Received 29 November 2012
Received in revised form 22 April 2013
Accepted 25 April 2013
Available online 2 July 2013

Keywords:
Computer simulation
High-energy-beam irradiation
Single-crystalline uranium dioxide
Molecular dynamics
Thermal spike model

ABSTRACT

The structural modification caused by the high-energy-ion irradiation of single-crystalline uranium dioxide was simulated by the molecular dynamics method. As the initial condition, high kinetic energy was supplied to the individual atoms within a cylindrical region of nanometer-order radius located in the center of the specimen. The potential proposed by Basak et al. [C.B. Basak, A.K. Sengupta, H.S. Kamath, J. Alloys Compd. 360 (2003) 210–216] was utilized to calculate interaction between atoms. The supplied kinetic energy was first spent to change the crystal structure into an amorphous one within a short period of about 0.3 ps, then it dissipated in the specimen. The amorphous track radius R_a was determined as a function of the effective stopping power gS_e, i.e., the kinetic energy of atoms per unit length created by ion irradiation (S_e: electronic stopping power, g: energy transfer ratio from stopping power to lattice vibration energy). It was found that the relationship between R_a and gS_e follows the relation $R_a^2 = a\ln(gS_e) + b$. Compared to the case of Si and β-cristobalite single crystals, it was harder to produce amorphous track because of the long range interaction between U atoms.

1. Introduction

Irradiation with heavy ions with energy range 100 MeV – few GeV have crucial effect on the structure and properties of various inorganic materials. Especially it is a tremendous feature of irradiation of heavy ions that high density electronic excitation causes movement of atoms and large irradiation effect in the material. For UO_2 material, addition of different oxides such as Gd_2O_3 causes refinement of crystal grains, lowering melting point and worsen the toughness [1]. The effect of high density electronic excitation depends largely on the initial structure and properties of the material. For example, low thermal conductivity and small crystal grain lead to localization of energy of electronic excitation and therefore to large effect of electronic excitation. So it can be considered that the irradiation effect of fission products with high energy during burning of nuclear fuels largely differs with and without addition of different oxides, i.e., burnable poison.

In the present study, as the basic research for development of next generation nuclear fuels with high burning ratio, the irradiation effect of heavy ions with high energy was studied by computer experiment. We simulated the structural modification after the addition of the initial excess energy (the kinetic energy of atoms) to the central region of UO_2 single crystal by the molecular dynamics method (MD). The threshold energy to produce amorphous track from atomic structure and radial distribution of atoms obtained from the present simulation.The objectives of our simulations are:

(i) To clarify the details of the high-energy dissipation process following the structural change of the irradiated nanoscale region for a UO_2 single crystal.
(ii) To determine the correlation between the initial excess energy given to the specimen and the radius of the track produced by an ion.

The obtained results were compared with those of computer experiments for Si and β-cristobalite performed by us previously [2].

2. Method of simulation

2.1. Molecular dynamics

The U and O atoms were arranged to form fluorite structure such that U atoms form face centered structure and O atoms sit at the center of the tetragonal cluster formed by U–U nearest

* Corresponding author at: Department of Materials Science and Engineering, Faculty of Engineering, Ibaraki University, 4-12-1 Nakanarusawa, Hitachi 316-8511, Japan. Tel.: +81 (294) 38 5074; fax: +81 (294) 38 5226.
E-mail address: sasajima@mx.ibaraki.ac.jp (Y. Sasajima).

0168-583X/$ - see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.nimb.2013.04.068

neighbor bonds. The bond length between U and O was 0.27 nm. The unit cell of fluorite structure was repeated five times each in ⟨100⟩,⟨010⟩ and ⟨001⟩ directions. The dimension of the calculation region was 2.74 nm × 2.74 nm × 2.74 nm and the number of atoms was 1500 in total. At first, the initial structure was relaxed by molecular dynamics method at 298 K. Then, the initial excess energy (the kinetic energy of atoms) was given in the cylindrical region with radius 0.25–0.5 nm in the central part of the calculation region. Hereafter we call this region as "center". The direction of the cylinder, i.e., direction of irradiated beam was taken to be ⟨001⟩. The initial condition, that cylindrical region with high kinetic energy appears, is corresponded to the assumption of thermal spike model. This condition also matches the experimental observation that cylindrical track is observed after ion irradiation. The excess energy is considered to be a part of energy deposited by irradiation of a heavy ion. At first, a heavy ion penetrates into the solid sample and loses its energy. The energy loss is called as stopping power, S_e, of which unit can be expressed in energy per unit length, such as keV/nm. The energy loss mainly changes in electronic excitation, however, the part of it elevates the kinetic energy of atoms in the cylindrical region around the heavy ion pass. We define this energy as effective stopping power, which is the excess kinetic energy of atoms created from stopping power

Table 1
Potential parameters for UO_2[4].

Parameters	O–O Pair	U–U Pair	U–O Pair
z_i	1.44	2.88	−5.76
a_i [Å]	3.82	5.76	−2.88
b_i [Å]	0.327022	0.327022	0.327022
c_i [kJ mol^{-1} Å^{-6}]	381	0	0
D_{ij} [Å]	0	0	13.6765
β_{ij} [Å^{-1}]	0	0	1.65
r^*_{ij} [Å]	0	0	2.369
f_0 [kJ mol^{-1} Å^{-1}]		4.07196	

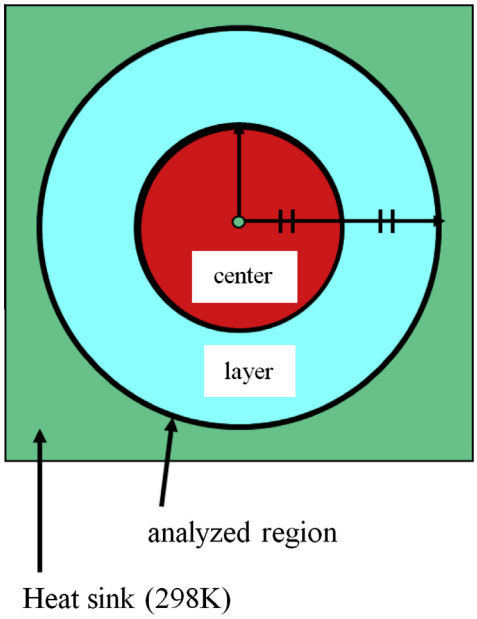

Fig. 1. Schematic of the analyzed region, center and layer. "Center" is the same as the central region supplied initial excess energy explained in the text, and "layer" is its peripheral region of which radius is two times larger than that of center.

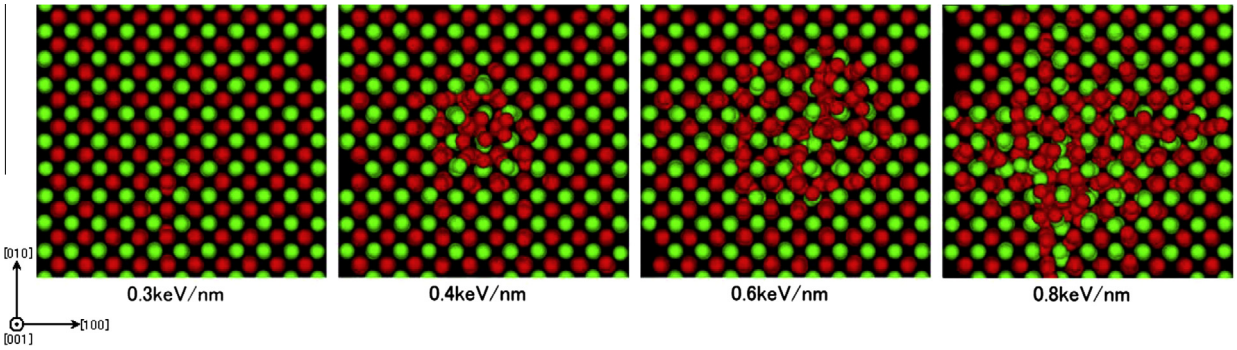

Fig. 2. Atomic structure of UO_2 at 3.00 ps after the supply of high kinetic energy. The effective stopping power for UO_2 is 0.30, 0.40, 0.60 and 0.80 keV/nm from left to right. The radius of the central region was 0.5 nm. Green and red circles represent U and O atoms, respectively.(For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.).

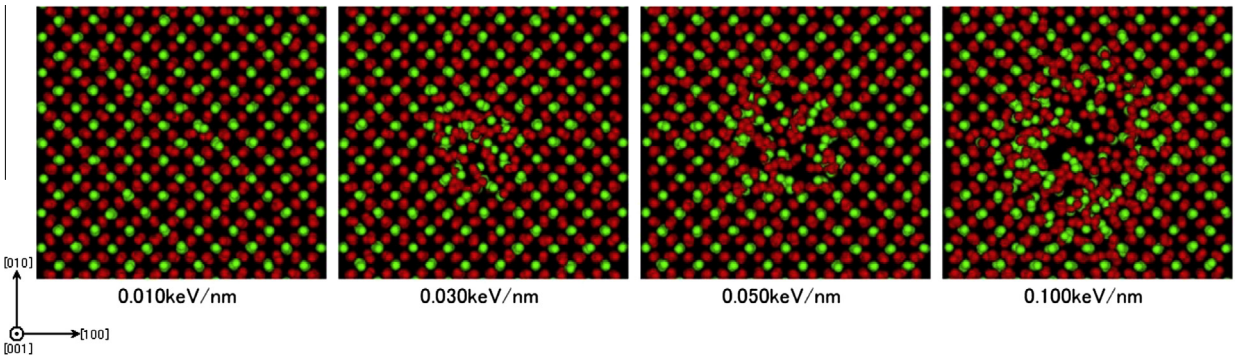

Fig. 3. Atomic structure of SiO_2 at 3.00 ps after the supply of high kinetic energy. The effective stopping power for SiO_2 is 0.010, 0.030, 0.050 and 0.100 keV/nm from left to right. The radius of the central region was 0.5 nm. Green and red circles represent Si and O atoms, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)