Dislocation Loops in Proton Irradiated Uranium-Nitrogen-Oxygen System

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Abstract

In this study, we investigated the dislocation loop types formed in the proton-irradiated uranium-nitrogen-oxygen (U-N-O) system, which involves uranium mononitride (UN), uranium sesquinitride (α-U_2N_3), and uranium dioxide (UO_2) phases. The dislocation loop formation is examined using specimens irradiated at 400°C and 710°C. Based on the detailed transmission-based electron microscopy characterization with i) the morphology-based on-zone and ii) the invisibility-criterion based two-beam condition imaging techniques, only a single type of dislocation loop in each phase is found: $a/2\langle110\rangle$, $a/2\langle111\rangle$, or $a/3\langle111\rangle$ dislocation loops in UN, α-U_2N_3, and UO_2 phases, respectively. Molecular statics calculations for the formation energy of perfect and faulted dislocation loops in UN phases indicate a critical loop size of ~ 6 nm, above which perfect loops are energetically favorable. This could explain the absence of faulted loops in the experimental observation of the irradiated UN phase at two temperatures. This work will enhance the understanding of irradiation induced microstructural evolution for uranium nitrides as advanced nuclear fuels for the next-generation nuclear reactors.

Keywords: radiation effects; dislocation loops; uranium nitride; nuclear fuels

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1. Introduction

Uranium mononitride (UN) has been considered a candidate fuel material for advanced nuclear systems including liquid metal-cooled fast reactors, high-temperature gas-cooled reactors, and light water reactors [1–10] due to its multiple advantages [3]: (i) higher fissile density (40% more uranium in UN than in uranium dioxide, UO_2), (ii) higher thermal conductivity, (iii) easier preprocessing, (iv) good irradiation stability and chemical compatibility with most potential cladding materials, and (v) longer fuel cycle time.

The phase and dislocation loop evolution in the uranium-nitrogen-oxygen system under proton irradiation has been recently studied [9]. Irradiation can enhance the oxidation of UN, which creates two fully coherent phases, alpha uranium sesquinitride (α-U_2N_3) and UO_2 [9]. UN has a rocksalt crystal structure, α-U_2N_3 has a body-center-cubic (BCC) based Mn_2O_3-type crystal structure [11], and UO_2 has a fluorite crystal structure [12]. The dislocation loop size was found three times larger in the two nitride phases, UN and α-U_2N_3, than in UO_2, while the number density was one order of magnitude higher in UO_2 than in the nitride phases [9].

However, the characteristics of dislocation loops such as the Burgers vector and habit plane in the abovementioned three phases under proton irradiation have not been studied in detail. Although the defects in actinide nitrides are rarely investigated, zirconium nitride (ZrN) was used as a common non-radioactive surrogate for UN to study the radiation effects [13,14] because ZrN shares the same crystal structure with UN. Yang et al. [13] used the high-resolution transmission electron microscopy (HRTEM) technique to show that vacancy-type pyramidal dislocation loops formed in ZrN under proton irradiation to 0.75 dpa at 800°C, while faulted dislocation loops were not observed using rel-rod dark field imaging technique. Bao et al. [15] showed a/2⟨110⟩ dislocation loop formation in the polycrystalline ZrN under gold irradiation. In another study, Jiao

24 et al. [14] observed both interstitial- and vacancy-types of dislocation loops using HRTEM in iron

25 irradiated nanocrystalline ZrN films.

26 Dislocation loop analysis in the traditional nuclear fuel UO_2 under heavy ion irradiation (using

27 Cs, Kr, or Xe ions) has been studied for decades. These studies [16–19] showed the exclusive

28 existence of $a/2\langle110\rangle$ perfect type of dislocation loops in heavy ion irradiated UO_2, while very

29 limited neutron or electron irradiation experimental data [20] showed the existence of only

30 $a/3\langle111\rangle$ faulted dislocation loops. The disparity is likely a result of cascade size difference

31 induced by heavy ions with varying mass and charge; this hypothesis may be validated using light-

32 ion irradiation, such as protons. In addition, molecular dynamics (MD) simulation studies of UO_2

33 [21–23] indicated that interstitial clusters may initially aggregate to form small $a/3\langle111\rangle$ faulted

34 dislocation loops, which is a more stable configuration compared to $a/2\langle110\rangle$ perfect dislocation

35 loops. With increasing size, unfaulting can occur due to the increasing stacking fault energy, which

36 has been reported by experiments and simulations in irradiated face-center-cubic (FCC) materials

37 [24–32]. Besides the direct investigation of irradiated UO_2, dislocation loop analysis of ion

38 irradiated thorium dioxide (ThO_2), sharing the same crystal structure of UO_2, has also been studied.

39 Mixed data have been reported [33–35] in irradiated ThO_2 with the formation of $a/2\langle110\rangle$ and/or

40 $a/3\langle111\rangle$ type of dislocation loops due to different irradiation conditions such as ion species,

41 temperature, and damage level.

42 To analyze proton-irradiation-induced dislocation loops in the U-N-O system (three phases:

43 UN and α-U_2N_3, and UO_2), we used developed scanning/transmission electron microscopy

44 (S/TEM) techniques with the aid of several methods for classification of loop types based on

45 observed contrast and morphology [36–38]. The on-zone STEM technique coupled with simulated

46 morphology maps has been recently developed and applied to characterize dislocation loop

3

47 formation in irradiated FCC [38] and BCC [36,37] alloys. It has been demonstrated that dislocation

48 loops of various types [29,38–40] in these alloys can be unambiguously identified, given the

49 correct orientation is used, based on their projection morphology when the dislocation loop size is

50 above a given size threshold, typically around 5-10 nm for most STEM instruments and imaging

51 conditions. In addition, STEM imaging has multiple advantages of (i) suppressing background

52 contrast to improve signal-to-noise ratio [41], (ii) exhibiting all dislocation loops within the TEM

53 specimen when the on-zone condition is satisfied [42], and (iii) the applicability of some common

54 conventional transmission electron microscopy (CTEM) techniques for dislocation loop analysis

55 such as the two-beam condition imaging that utilizes the $\vec{g} \bullet \vec{b}$ invisibility criterion [43,44].

56 Furthermore, to gain a fundamental understanding towards the observed dislocation loop types,

57 MD simulations are applied to reveal the energetically favorable atomic configuration and

58 calculate the size-dependent formation energy of dislocation loops in irradiated UN.

59 **2. Methods**

60 2.1 Proton Irradiation

61 The UN-5wt.%UO$_2$ samples were irradiated with 2 MeV proton ions to reach the total fluence

62 of 8×10^{18} ions/cm^2 at 400°C and 710°C, respectively [9]. The proton flux was maintained at

63 5.2×10^{13} ions/(cm^{2}s) for the low-temperature irradiation and 7.5×10^{13} ions/(cm^{2}s) for the high-

64 temperature irradiation, resulting in a damage rate of 4.1×10^{-6} and 5.9×10^{-6} dpa/s, respectively for

65 UN. The Stopping and Range of Ions in Matter (SRIM) 2013 code in "Detailed Calculation with

66 Full Damage Cascades" mode [45] was used to calculate the proton-irradiation damage with the

67 damage profiles for UN, α-U$_2$N$_3$ and UO$_2$ shown previously [9]. The region of interest of below 3

68 μm is located at the common plateau portion of the damage curves of three phases, where the

69 damage dose is about 0.5~0.6 dpa for the three phases. Based on the SRIM results, the calculated

70 dose varied by only 10% across the three phases, which is within the variance expected for a single

71 phase across the depth region investigated. This depth range was selected with the purpose of

72 avoiding significant injected-proton-interstitial effects [46,47].

73 2.2 Microstructural Characterization

74   After the ion irradiation, the "lift-out" technique using focused ion beam (FIB) equipped on

75 an FEI Quanta 3D scanning electron microscope was conducted to prepare electron transparent

76 specimen, followed by low-energy (5 kV and 2 kV) ion sputtering to reduce FIB-induced damages

77 on the sample surface. Three phases, UN, α-U_2N_3, and UO_2 after irradiation at two temperatures,

78 were identified and located by performing energy-dispersive X-ray spectroscopy (EDS) mapping

79 using a Titan Themis 200 Scanning and Transmission Electron Microscope (S/TEM). Detailed

80 chemical mapping results with limited strain-based contrast imaging have been reported in the

81 previous study [9].

82   Dislocation loop characterization and Burgers vector analysis for the two nitride-based phases

83 were conducted with two techniques: (i) two-beam condition imaging in STEM mode using the

84 $\vec{g} \cdot \vec{b}$ invisibility criterion [48] involving multiple tilting conditions, and (ii) the recently

85 developed on-zone STEM imaging [36–38]. The diffraction patterns were obtained after the

86 sample tilting using CTEM, and then STEM was used for imaging at the selected tilting conditions.

87 The zone axes used for all the dislocation loop characterization were one of the commonly used

88 major low-index ones including [001], [011], and [111] because the dislocation loop morphology

89 maps are available in the literature for the FCC (similar to UN) and BCC (similar to α-U_2N_3)

90 crystal structures, and the $\vec{g}$ vectors near these zone axes were selected to be low-order as well.

91   Dislocation loop characterization in proton irradiated UO$_2$ at 710°C was conducted using the

92    $\vec{g} \cdot \vec{b}$ invisibility criterion in CTEM mode. The characterization for UO$_2$ irradiated at 400°C was

93   not conducted because dislocation loops are too small (e.g., < 5 nm) to be identified using S/TEM

94   techniques, especially with the existence of possible FIB-induced black-dot damage.

3. Results

96   3.1 Dislocation Loops in UN after Irradiation at 400°C and 710°C

97   Figure 1 shows the dislocation loop structures in the UN phase after irradiation at 400°C. The

98   on-zone [001] STEM bright-field (STEM-BF) image exhibits all dislocations and dislocation loops,

99   while kinematic two-beam condition STEM-BF images utilizing $\vec{g} \cdot \vec{b}$ invisibility criterion reduce

100   or eliminate the contrast of some of the dislocation loops. Multiple $\vec{g}$ conditions are achieved near

101   the [001] zone axis, including $\vec{g}_{\bar{2}00}$, $\vec{g}_{\bar{2}20}$, $\vec{g}_{020}$ and $\vec{g}_{\bar{2}\bar{2}0}$. The dislocation loop morphology map

102   developed for irradiated FCC-based alloys at [001] zone axis (adapted from Ref. [38]) is shown in

103   Figure 1 as well. Because the rocksalt crystal structure of UN is similar to the FCC crystal structure,

104   similar types of dislocation loops, i.e., a/2⟨110⟩{110} perfect and a/3⟨111⟩{111} faulted loops, if

105   exist, are expected to form under irradiation.

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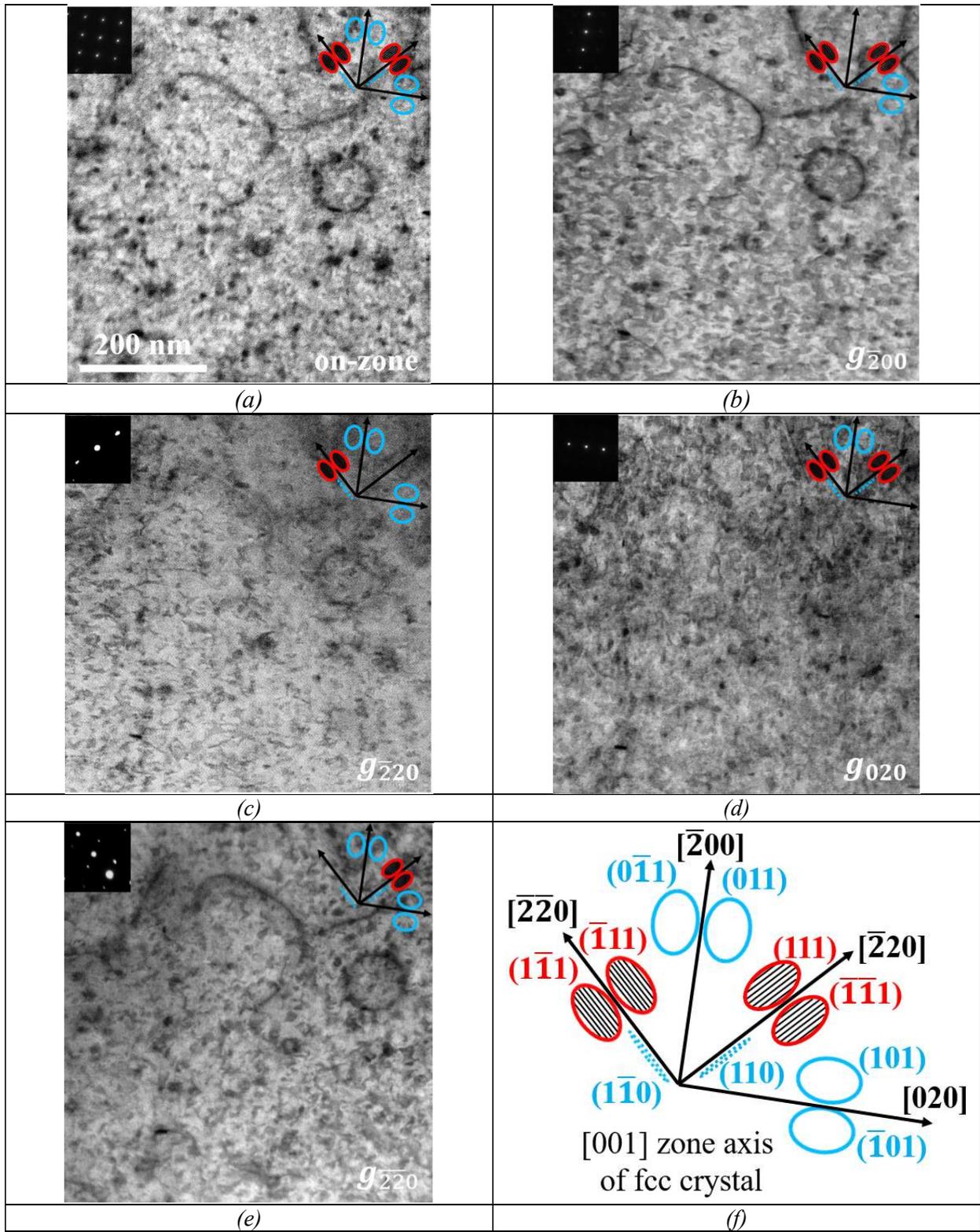
Figure 1. STEM-BF images of irradiated UN phase at 400°C with imaging conditions of (a) on-zone [001], as well as kinematic two-beam conditions with (b) $\vec{g}_{\bar{2}00}$, (c) $\vec{g}_{\bar{2}20}$, (d) $\vec{g}_{020}$ and (e) $\vec{g}_{\bar{2}\bar{2}0}$, respectively. The morphology and interior contrast of the visible dislocation loops is shown at the top right corner for each imaging condition, based on the developed dislocation loop morphology map

developed for FCC based alloys at [001] zone axis (adapted from Ref. [38]). The full crystallographic orientation of all the dislocation loop type is shown in (f), with its orientation rotated and correlated to the STEM images in (a)~(e). Note that the a/3⟨111⟩ faulted loops are not observed in the microstructures.

107

108   As can be seen, there is no a/3⟨111⟩ faulted dislocation loop observed in irradiated UN at the

109   temperature of 400°C, because (i) the interference shadow contrast would have been exhibited if

110   faulted loops exist in irradiated or quenched FCC alloys [29,38,49–52] due to the inserted faulted

111   plane causing a phase change of the electron wave [48,53], and (ii) no loops appear or disappear

112   in the same way as the faulted dislocation loops in the STEM images of different two-beam

113   conditions that is determined by the $\vec{g} \cdot \vec{b}$ invisibility criterion. Therefore, all observed dislocation

114   loops are considered to be perfect-type in the irradiated UN, assuming all faulted loops would

115   appear non-edge-on at the [001] viewing direction.

116   The irradiation-induced dislocation loop structures at 710°C are shown in Figure 2, with the

117   on-zone [$\bar{1}$11] STEM-BF image exhibits all dislocations and dislocation loops, as well as three

118   kinematic two-beam condition STEM-BF images near the [$\bar{1}$11] zone axis with $\vec{g}_{\bar{2}0\bar{2}}$, $\vec{g}_{02\bar{2}}$ and

119    $\vec{g}_{\bar{2}\bar{2}0}$, respectively. The dislocation loop morphology map developed for irradiated FCC-based

120   alloys at [$\bar{1}$11] zone axis (adapted from Ref. [38]) is shown in Figure 2 as well. Again, only the

121   perfect dislocation loops in the microstructure exist after the proton irradiation at 710°C.
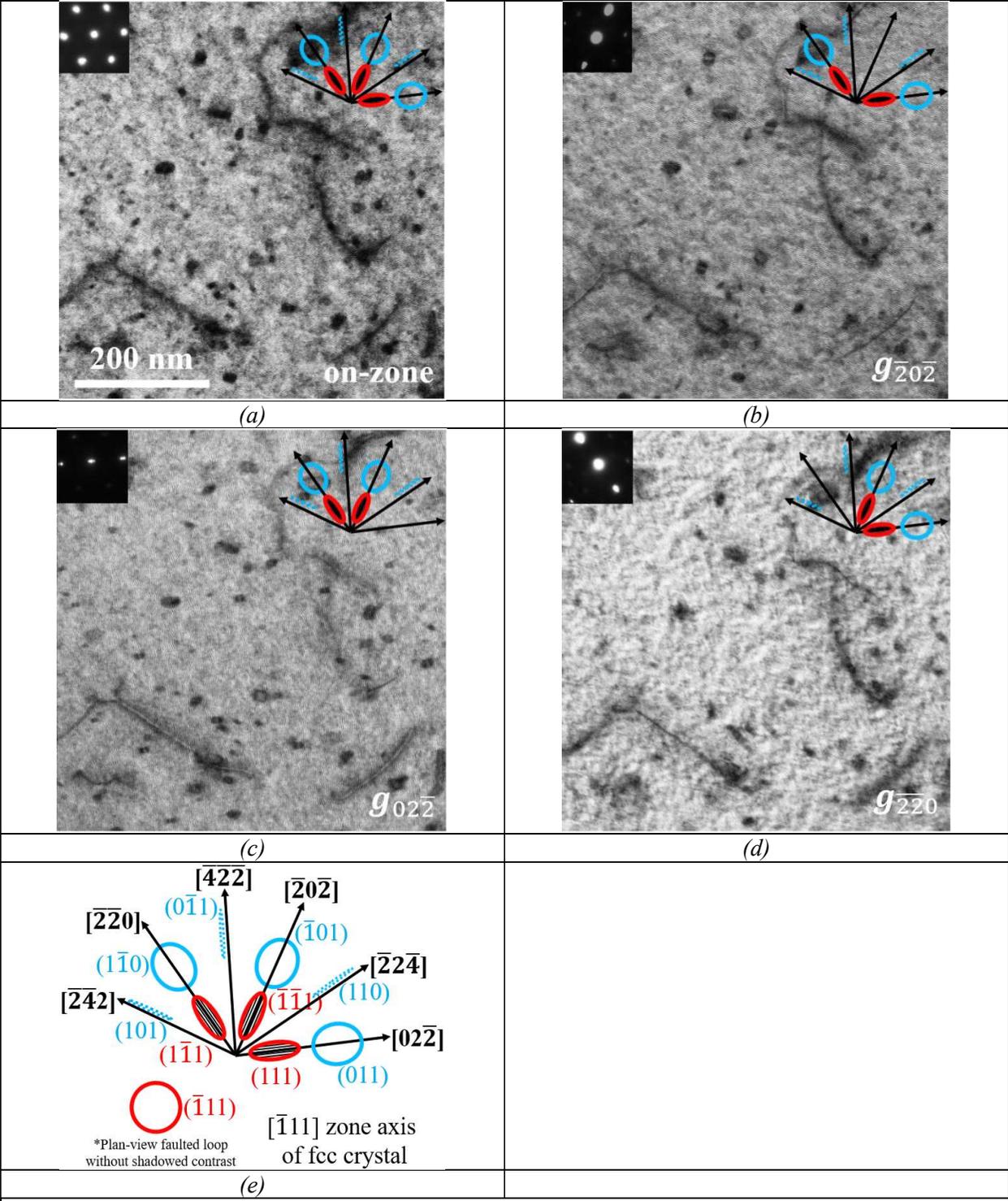
Figure 2. STEM-BF images of irradiated UN phase at 710°C with imaging conditions of (a) on-zone [$\bar{1}$11], as well as kinematic two-beam conditions with (b) $\vec{g}_{\bar{2}0\bar{2}}$, (c) $\vec{g}_{02\bar{2}}$ and (d) $\vec{g}_{\bar{2}\bar{2}0}$, respectively. The morphology and interior contrast of the visible dislocation loops is shown at the top right corner for each imaging condition, based on the developed dislocation loop morphology map developed for FCC based alloys at [$\bar{1}$11] zone axis (adapted from Ref. [38]). The full crystallographic orientation of all the dislocation loop type is shown in (e), with its orientation rotated and correlated to the STEM images in (a)~(d). Note that the a/3⟨111⟩ faulted loops are not observed in the microstructures.

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3.2 Dislocation Loops in α-U$_2$N$_3$ after Irradiation at 400°C and 710°C

The dislocation loop structures of proton irradiated α-U$_2$N$_3$ at 400°C are shown in Figure 3, with the on-zone [101] STEM-BF image exhibits all dislocations and dislocation loops, as well as three kinematic two-beam condition STEM-BF images near the [110] zone axis with $\vec{g}_{040}$, $\vec{g}_{40\bar{4}}$, $\vec{g}_{\bar{2}22}$ and $\vec{g}_{22\bar{2}}$, respectively. The dislocation loop morphology map developed for irradiated BCC Fe-based alloys at [101] zone axis (adapted from Ref. [36]) is shown in Figure 3(f). Because the Mn$_2$O$_3$ crystal structure of the α-U$_2$N$_3$ phase is similar to the BCC crystal structure, and thus, similar types of dislocation loops, i.e., a $\langle 100 \rangle$ {200} and a/2$\langle 111 \rangle${111}, if exist, are expected under irradiation.

(a)	(b)

Figure 3. STEM-BF images of irradiated α-U₂N₃ phase at 400°C with imaging conditions of (a) on-zone [101], as well as kinematic two-beam conditions with (b) $\vec{g}_{040}$, (c) $\vec{g}_{40\bar{4}}$, (d) $\vec{g}_{\bar{2}22}$ and (e) $\vec{g}_{22\bar{2}}$, respectively. The morphology of the visible dislocation loops is shown at the top right corner for each imaging condition, based on the developed dislocation loop morphology map developed for BCC based alloys at [101] zone axis (adapted from Ref. [36]). The full crystallographic orientation of all the dislocation loop type is shown in (f), with its orientation rotated and correlated to the STEM images in (a)~(e). Note that the a⟨100⟩ loops are not observed in the microstructures.

132

133   By carefully examining the visibility of dislocation loops under all the selected two-beam

134   conditions, as well as their morphology prediction, it is determined that only a/2⟨111⟩type of

135   dislocation loops exist in the irradiated α-U₂N₃ at 400°C, while a ⟨100⟩ loops are not observed.

136   In addition, all the dislocations are also identified with the type of a/2⟨111⟩.

11

137   At a higher temperature of 710°C, the proton irradiation-induced dislocation loop structures

138   are shown in Figure 4 in corresponding STEM images. In this case again, only the a/2⟨111⟩ type

139   dislocation loops are identified from the elliptical shape and the direction of the major axis

140   observed, and by the comparison with the dislocation loop morphology map in Figure 4(f).

(a)   (b)

(c)   (d)
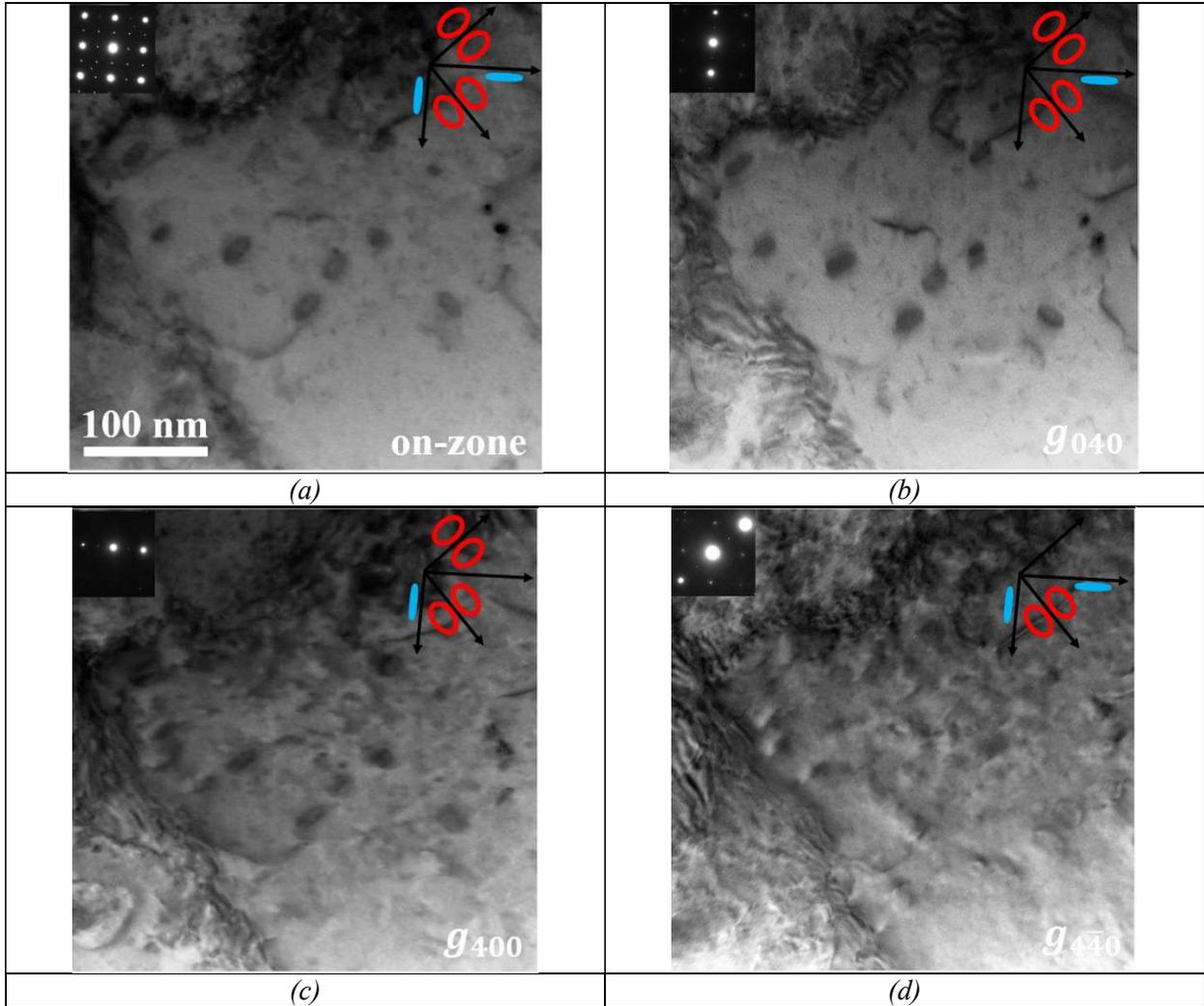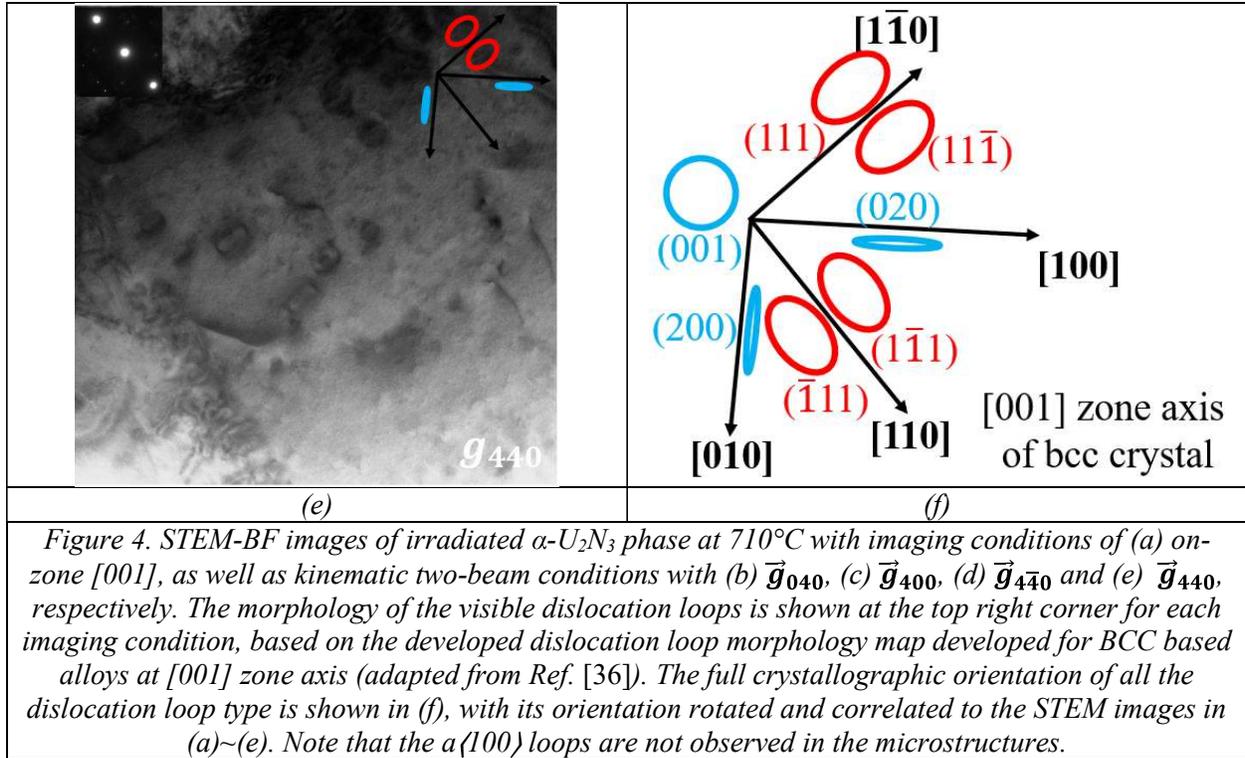
Figure 4. STEM-BF images of irradiated α-U$_2$N$_3$ phase at 710°C with imaging conditions of (a) on-zone [001], as well as kinematic two-beam conditions with (b) $\vec{g}_{040}$, (c) $\vec{g}_{400}$, (d) $\vec{g}_{4\bar{4}0}$ and (e) $\vec{g}_{440}$, respectively. The morphology of the visible dislocation loops is shown at the top right corner for each imaging condition, based on the developed dislocation loop morphology map developed for BCC based alloys at [001] zone axis (adapted from Ref. [36]). The full crystallographic orientation of all the dislocation loop type is shown in (f), with its orientation rotated and correlated to the STEM images in (a)~(e). Note that the a⟨100⟩ loops are not observed in the microstructures.

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142 3.3 Dislocation Loops in UO$_2$ after Irradiation at 710°C

143   The dislocation loop structures of proton irradiated UO$_2$ at 710°C are shown in Figure 5, with

144 the Burgers vector analysis conducted by using the $\vec{g} \cdot \vec{b}$ invisibility criterion in the CTEM mode.

145 This technique was chosen for loop type analysis in irradiated UO$_2$ over the previously mentioned

146 morphology-based technique, because as can be seen in Figure 5, the dislocation loop size is quite

147 small (< 5 nm), making the morphology ambiguous to identify.

148   Figure 5 shows CTEM-BF images of UO$_2$ phase obtained using various g conditions near [001]

149 zone axis, including $\vec{g}_{200}$, $\vec{g}_{220}$ and $\vec{g}_{2\bar{2}0}$. Table 1 shows invisibility criteria for dislocation loops

150 with four variants of a/3⟨111⟩ faulted type and six variants of a/2⟨110⟩ perfect type Burgers vector

151 directions under the three selected $\vec{g}$ two-beam conditions. In Figure 5, two different families of

152 dislocation loops were identified by letters A and B and colors red and yellow, respectively. Family

13

153   A dislocation loops were visible at $\vec{g}_{200}$ and $\vec{g}_{220}$, while became invisible at $\vec{g}_{2\bar{2}0}$ as shown in

154   Figure 5(a), 5(b) and 5(c), respectively. Family B dislocation loops were visible at $\vec{g}_{200}$ and $\vec{g}_{2\bar{2}0}$

155   while disappears at $\vec{g}_{220}$ as shown in Figure 5(a), 5(b) and 5(c), respectively. Table 1 also lists

156   possible Burgers vector directions for loop family A and B based on their appearances and

157   disappearances at given $\vec{g}$ conditions. Burgers vector direction of loop family A was either [111]

158   or [$\bar{1}\bar{1}1$] or [110], while Burgers vector direction of loop family B was either [$1\bar{1}\bar{1}$] or [$1\bar{1}1$] or

159   [$\bar{1}10$], as shown in Table 1. It should be noted that current $\vec{g}$ conditions identify both loop families

160   A and B as either a/3⟨111⟩ type faulted dislocation loops or a/2⟨110⟩ type perfect dislocation loops.
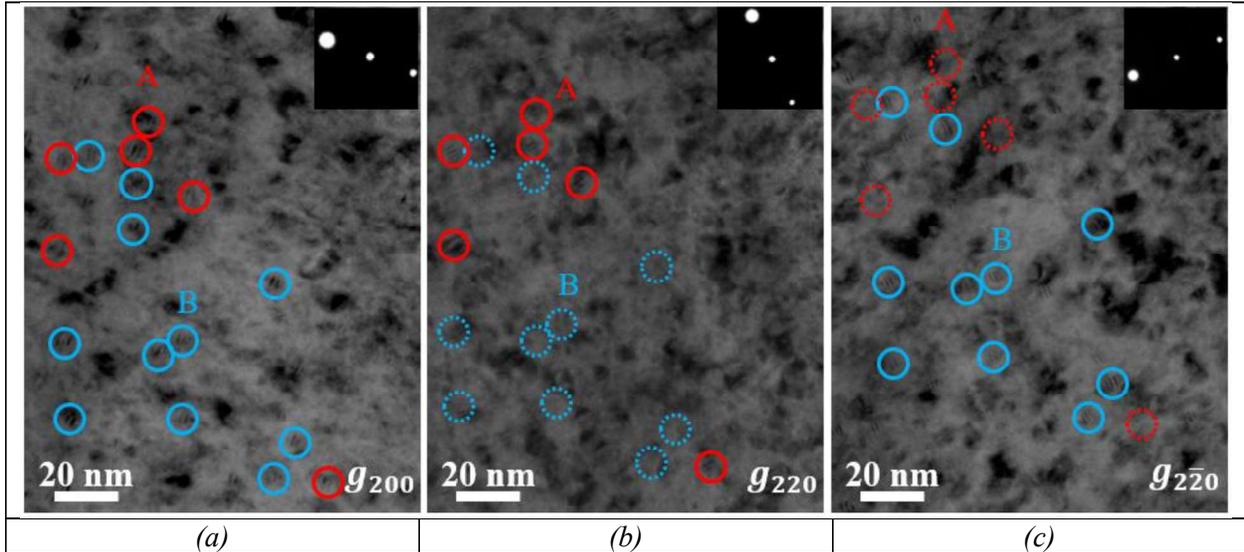
Figure 5. CTEM-BF images of irradiated UO2 phase at 710°C with imaging conditions of kinematic two-beam conditions with (a) $\vec{g}_{200}$, (b) $\vec{g}_{220}$, and (c) $\vec{g}_{2\bar{2}0}$ near the [001] zone axis. Note that the dislocation loop size in the irradiated UO_2 phase is so small that the morphology-based technique mentioned before may not readily apply due to the ambiguous observed dislocation loop morphology.

161

162   **Table 1.** Burgers vector analysis using $\vec{g} \cdot \vec{b} = 0$ invisibility criterion for the proton irradiated UO_2 phase
163   at 710°C.

$\vec{b}$	$\vec{g}$ (zone axis = [001])			Identified loops
	[200]	[220]	[$2\bar{2}0$]	
a/3[111]	V	V	I	A ⭕
a/3[$\bar{1}\bar{1}1$]	V	V	I	A ⭕
a/3[$1\bar{1}\bar{1}$]	V	I	V	B ⭕
a/3[$1\bar{1}1$]	V	I	V	B ⭕

14

a/2[011]	I	V	V	–
a/2[101]	V	V	V	–
a/2[110]	V	V	I	A
a/2[0$\bar{1}$1]	I	V	V	–
a/2[10$\bar{1}$]	V	V	V	–
a/2[$\bar{1}$10]	V	I	V	B ○

164

165 To further confirm the dislocation loop type, high magnification CTEM-BF and weak beam

166 dark field (WBDF) images of dislocation loops in the irradiated UO_2 phase at $g = [2\bar{2}0]$ near $[001]$

167 zone axis are shown in Figure 6(a) and 6(b), respectively. It is seen that dislocation loops have

168 elliptical shape with stacking fault fringes inside of it. Since only faulted a/3⟨111⟩ type dislocation

169 loops can contribute the inserted extra plane that causes faulted stacking sequence, it can be

170 concluded that loop family A and loop family B are of different variants, but both are a/3⟨111⟩

171 type faulted dislocation loops. This observation is in good agreement with the proton irradiation

172 ThO_2 in the previous study [34], where only a/3⟨111⟩ type of faulted dislocation loops were found.
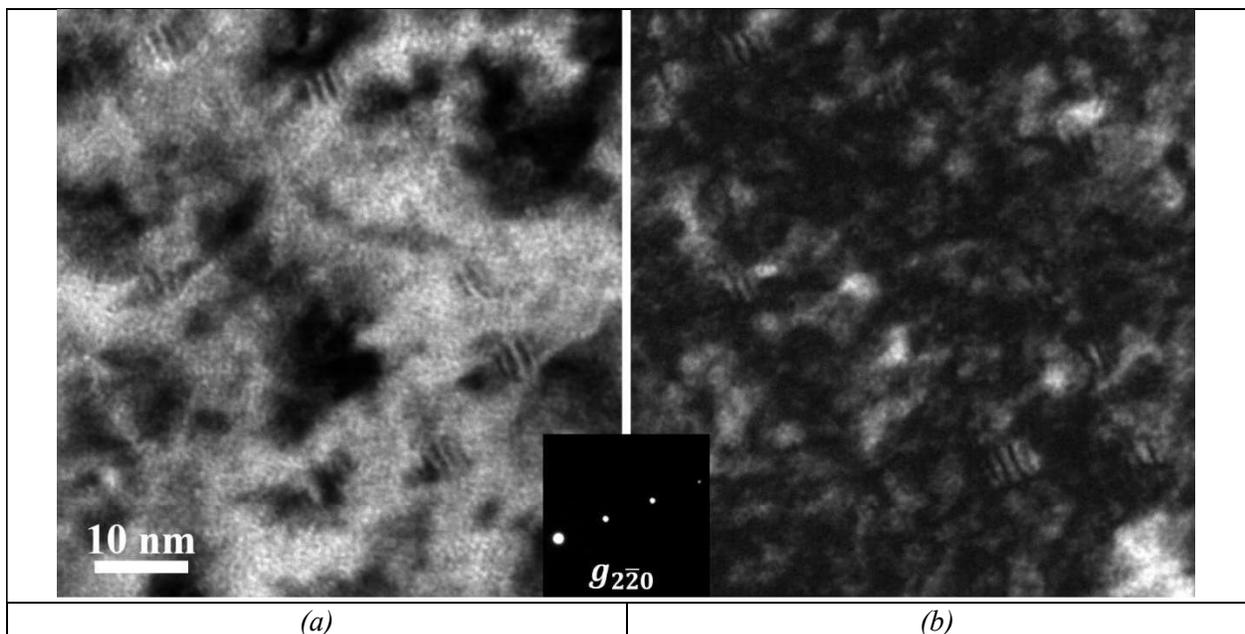
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| (a) | (b) |

Figure 6. CTEM- (a) BF and corresponding (b) weak beam dark field (WBDF) images of UO2 phase irradiated at 710°C obtained using $\vec{g}_{2\bar{2}0}$ near the [001] zone axis.

4. Discussion

To examine the structure and energetics of the two types of dislocation loops in UN, we applied molecular statics (MS) calculation using the LAMMPS package (https://lammps.sandia.gov) [54]. The interatomic potential considering the angular dependence was developed by Tseplyaev and Starikov [55], based on force-matching to DFT database. This potential was demonstrated to have a low fitting error and well-reproduced thermodynamic properties for the B1-phase of UN [55]. Therefore, this potential is utilized. Note that it does not explicitly consider charge property, which may be justified by the fact that the bonding between U and N atoms is metallic-like [56].

In the crystal structure of B1-phase, there are predominantly two types of dislocation loops formed under irradiation, i.e. a/3⟨111⟩ faulted loops and a/2⟨110⟩ perfect loops. Notably, the faulted loops have been reported in irradiated titanium carbide (TiC) [57,58], zirconium carbide (ZrC) [59] and high-entropy carbide [60]. Here, we consider pure-edge type a/3⟨111⟩ and a/2⟨110⟩ loops on {111} and {110} habit planes, respectively. To ease the construction of loops, perfect simulation cells with z-axis oriented in [111] (15.2 nm × 15.5 nm × 10.1 nm, 164,736 atoms) and [011] directions (15.6 nm × 15.2 nm × 10.3 nm, 168,960 atoms) are created and relaxed to ground state, respectively.

To study the energetics of perfect and faulted dislocation loops in UN, we need to first identify the atomic structure for the loops. For a/3⟨111⟩ loop, due to the electrostatic interaction, a single layer of {111} loop would be energetically prohibited. With double layer {111} loops, the stoichiometry can significantly reduce the energy penalty. In this case, there are a few possible

16

195　stacking sequences for the a/3⟨111⟩ loops, which are demonstrated in Figure 7. Figure 7(a) shows

196　the perfect stacking. By comparison, Figure 7(b-d) indicate two inserted atomic layers (U and N

197　layer) differently: "S" denotes normal stacking in the sublattice leading to a standard stack fault

198　and "OS" denotes overlapping stacking, which would be prohibited in an FCC structure, but can

199　be stabilized with interweaving atomic layers with an opposite charge. Using the same notation

200　for the insert layers, Figure 8 compares the formation energy of a loop with 3 nm in diameter (the

201　only difference lies in the stacking). Figure 7(d) corresponds to the lowest energy configuration.

202　With this stacking, the stacking fault energy (γ) is calculated to be 1.54 J/m^2. Also, such stacking

203　is used for the energetic calculations for faulted loops at different sizes. For a/2⟨110⟩ loop, the

204　double-layer {110} stacking is shown in Figure 9, with each layer containing an equal number of

205　interstitials of both atom types. After identifying the stacking, loops at different radii are created

206　in the shape of disk in the simulation box. Then the system is relaxed during an energy

207　minimization so that the system pressure tensor is reduced to zero; this is followed by multiple

208　rounds of static minimization with conjugate gradient method.
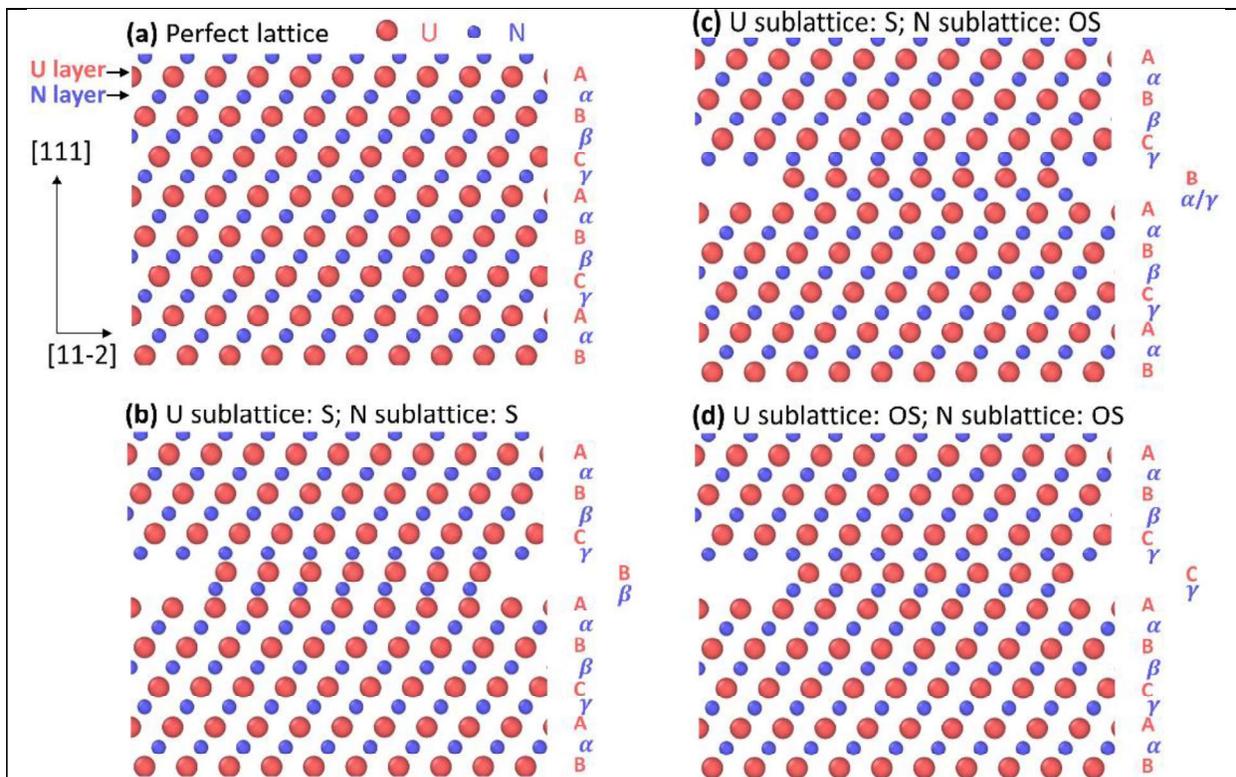
Figure 7. (a) Prefect lattice stacking; (c-d) different stacking for the inserted two layers (U and N layers, shape in disk). S: normal stacking in the sublattice; OS: overlapping stacking in the sublattice.

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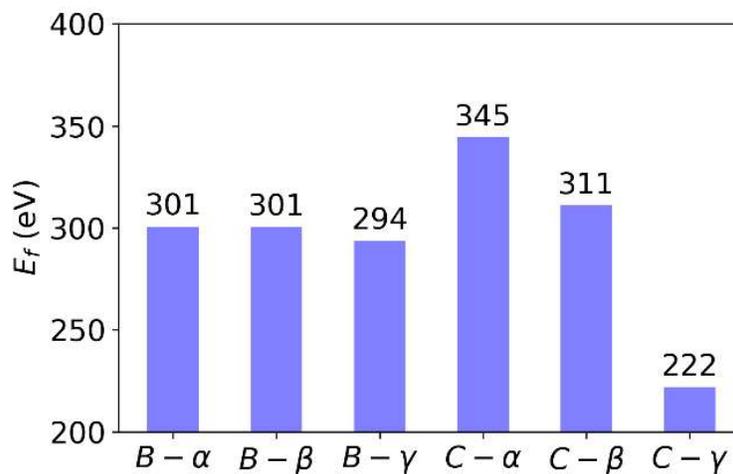
Figure 8. Formation energy of 3.0 nm diameter faulted loops for different stacking. The labels denote the stacking of the two insert atomic layers (see Figure 7b-d). Lowest energy (C-γ) corresponds to Figure 7(d).

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Figure 9. Lattice stacking of a perfect loop with two inserted atomic layers (shape in disk).

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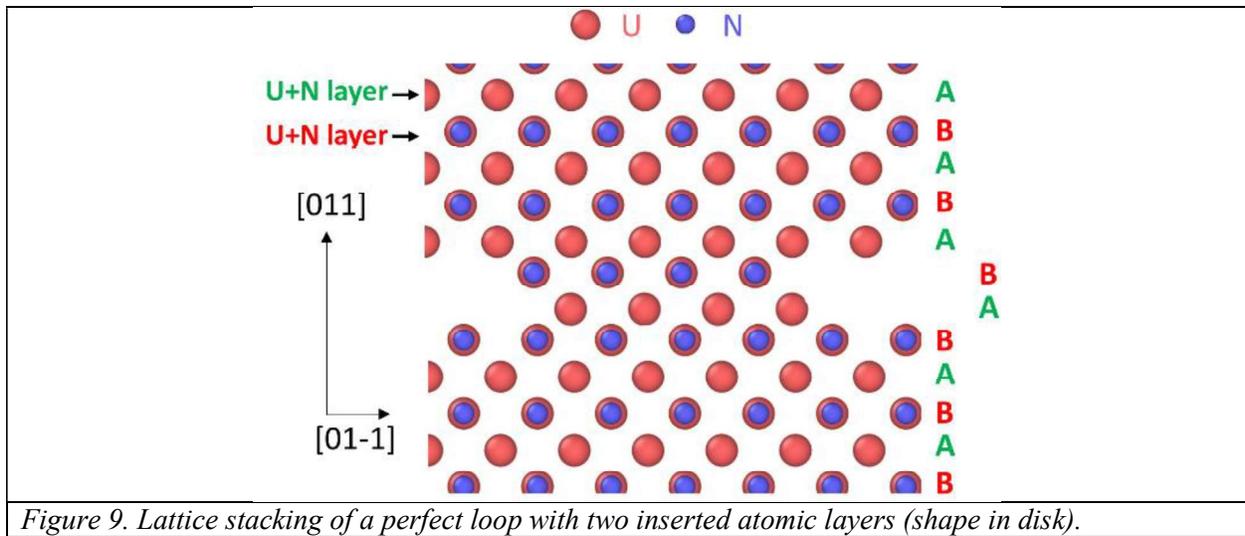
213   The loop formation energies are compared as a function of loop size as shown in Figure 10. It

214   is expected that the faulted loop becomes energetically less favorable as loop size increases,

215   because of the energy penalty from stacking fault [61]. As the loop size is below ~6 nm in diameter,

216   a/3⟨111⟩ extrinsic loop is slightly energetically favorable, while as the size becomes larger, there

217   is an energetic preference to transform to a prefect loop. This can be achieved by nucleating

218   Shockley partial dislocations [62] to initiate the unfaulting process. Note that due to the energy

219   barrier of dislocation nucleation and migration [62], one may observe large faulted loops,

220   especially at low temperatures without deformation. Although the average dislocation loop size is

221   6.2 ± 1.7 nm at 400°C [9], which is close to the critical size of ~6 nm calculated by MS simulation,

222   a/3⟨111⟩ faulted loops were not observed in this work. The observation indicates the unfaulting

223   process of dislocation loops in irradiated UN might happen at even smaller size. A similar

224   unfaulting process has been observed in neutron [63] and proton [59] irradiated ZrC, where ZrC

225   shares the same crystal structure as UN. Faulted dislocation loops formed with the average size of

226   4.3 ± 0.5 nm and 5.8 ± 0.6 nm in proton irradiated ZrC up to 0.7 dpa and 1.5 dpa, respectively, at

227   800°C [59], while only unfaulted perfect dislocation loops with the average size of 9.85 nm were

228   observed in proton irradiated zorch up to 2 dpa at 1125°C [64]. Therefore, to reveal the unfaulting

229   process in UN, proton irradiation experiment at lower temperature, i.e. <400°C, or lower dose,

230   needs to be pursued in future studies.

Figure 10. Formation energy E_{ve} of a/2⟨110⟩ and a/3⟨111⟩ dislocation loops at different radii (R).
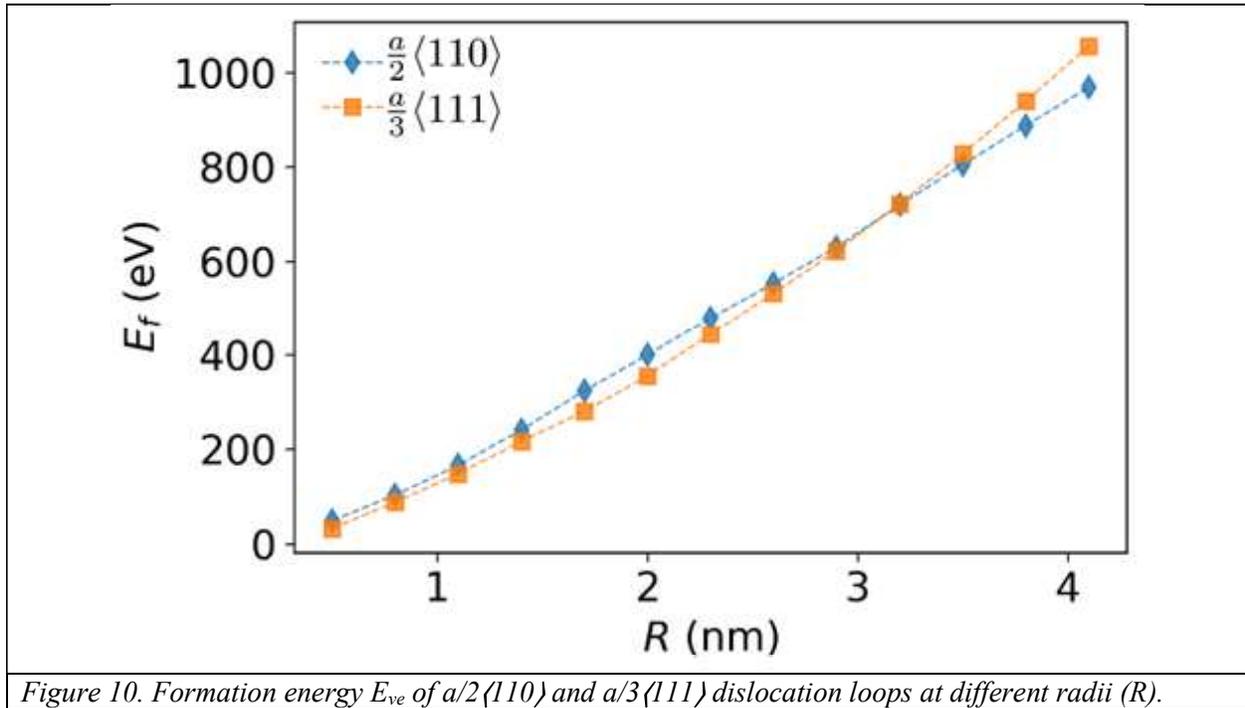
231

232   In this study, only a/2⟨111⟩ type of dislocation loops formed after proton irradiation at two

233   temperatures in α-U_2N_3 that has a Mn_2O_3-type BCC crystal structure. It has been extensively

234   reported in BCC-based ferritic alloys that perfect a/2⟨111⟩ and a⟨100⟩ loops can form under

235   irradiation [65–73]. It would have been interesting to compare the formation energy of these two

236   types of dislocation loops in α-U_2N_3 using MS calculation, but the interatomic potential of α-U_2N_3

237   has not been developed. Similar a/2⟨111⟩ perfect dislocation loop formation was observed in In_2O_3

238   [74] that has the same Mn_2O_3-type BCC crystal structure, where these loops formed from

239   misfitting inclusions at smaller misfit values than straight dislocations during In_2O_3 nanorod

240   growth rather than under irradiation. To fully understand the defect evolution under irradiation in

241 α-U_2N_3, the loop formation mechanism needs to be further explored via well-designed experiments

242 and modeling.

243   The dislocation loop type in proton irradiated UO_2 is in good agreement with that in proton

244 irradiated ThO_2, where only a/3⟨111⟩ faulted dislocation loops were found [34]. The average loop

245 size is quite small of 3.3 nm [9] and 4.5 nm [34] in proton irradiated UO_2 and ThO_2, respectively.

246 On the contrary, only a/2⟨110⟩ perfect loops of larger size were observed in heavy ion irradiated

247 UO_2 [17–19] and ThO_2 [35]. Similar to UN, MD calculations of loop formation energy in UO_2

248 [21,22] and ThO_2 [35] indicate that, a/3⟨111⟩ faulted dislocation loops are energetically favorable

249 when their size is less than a few nanometers, above which the unfaulting process may occur.

250   Irradiation induced defects may affect mechanical and thermal properties of ceramic nuclear

251 fuels and their non-radioactive surrogate, cerium dioxide (CeO_2) [75–81]. The thermal transport

252 behavior in CeO_2 is significantly affected by the type of dislocation loops, where perfect loops

253 only slightly reduce the thermal conductivity while faulted loops lead to an unusually large

254 reduction of thermal conductivity [77]. This was ascribed to extensive phonon scattering because

255 of a stronger strain field surrounding faulted loops. The effects of loop type and size on thermal

256 conductivity may also exist in UN, and more efforts on this topic are needed in the future.

5. Conclusion

258   The dislocation loop type analysis in proton irradiated UN and α-U_2N_3 phases at 400°C and

259 710°C, as well as irradiated UO_2 at 710°C are systematically investigated. It is revealed that a

260 single dislocation loop type of a/2⟨110⟩, a/2⟨111⟩ and a/3⟨111⟩ exists in UN, α-U_2N_3 and UO_2

261 phases, respectively. For the UN phase, MS simulation reveals that the formation energy of

262 a/3⟨111⟩ faulted loops is lower than a/2⟨110⟩ perfect loops when the dislocation loop size is smaller

263 than 6 nm, which is well below the resolution limit of the selected STEM imaging condition; it

264 explains why only perfect type of dislocation loops are observed in UN at two test temperatures.

265 This result is the first to report the single dislocation loop type formed in the proton irradiated

266 uranium nitrides and uranium dioxide phases, though future work concerning different radiation

267 conditions may be conducted to further investigate other potential dislocation loop type formation

268 in the systems.

269

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Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: