Mechanical Properties of UO₂ Thin Films under Heavy Ion Irradiation
using Nanoindentation and Finite Element Modeling

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Abstract

The mechanical response of UO₂ to irradiation is becoming increasingly important due to the shift to higher burn-up rates in the next generation of nuclear reactors. In the current study, thin films of UO₂ were deposited on YSZ substrates using reactive-gas magnetron sputtering. Nanoindentation was used to measure the mechanical properties of the as-grown and irradiated films. Finite element modeling was used to account for the substrate effect on the measurements. In order to study the effect of displacement cascades accompanying gas bubbles, 5000Å UO₂ films were irradiated with 600 keV Kr⁺ ions at 25°C and 600°C. These irradiation conditions were used to confine radiation damage effects and implanted gas within the film. Results showed an increase in the film hardness and yield strength with dose, while elastic modulus initially decreased with irradiation and then kept increasing with dose. The change in hardness and elastic modulus is attributed to the introduction of gas bubbles and displacement cascade damage. Irradiation at 600°C resulted in a decrease in the hardness and elastic modulus after irradiation using 600 keV Kr⁺ at a dose of 1E14 ions/cm². Both hardness and elastic modulus then increased with irradiation dose. This behavior is attributed to recrystallization during irradiation at 600°C and the formation of nanocrystallite regions with diameter and density that increase with dose. The calculation of the critical resolved shear stress (CRSS) demonstrated that nanocrystals are the primary cause for film hardening based on the Orowan hardening mechanism.

Keywords: UO₂, thin films, nanoindentation, mechanical properties, heavy ion irradiation

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1. Introduction

As the nuclear industry moves toward high fuel burn-up, there is a need for advancing our knowledge of the mechanical behavior of nuclear fuels to assure the fuel stability and durability. At the same time, pellet cladding interaction depends on the cladding mechanical properties as well as the level of stresses introduced in the fuel due to the fission gas release. Stresses within the pellet may induce cracks that depend on the fuel thermal expansion, thermal conductivity and the elastic properties [1 - 5].

Indentation techniques have been applied to study the effects of irradiation on the mechanical properties of UO_2 . Spino *et al.* used microhardness to the study of the change in elastic/plastic properties of UO_2 -LWR fuel pellets, irradiated in power reactor to a burn-up rate ranging from 40 to 100 GWd/t M [6]. The entire fuel rod hardness increased as the average burn-up increased up to 67 GWd/t M, but it decreased again as the average burn-up increased to 80 GWd/t M due to the healing of accumulated radiation damage. The periphery of the fuel rod softened and the depth of the rim zone increased with the average burn-up following the porosity build up profile [6]. In addition, material recrystallization at the pellet edge was observed [7, 8]. Kim *et al.* applied Knopp indentation and found similar behavior with neutron irradiation, that the UO_2 hardness increased with neutron dose up to a certain point then decreased [9, 10], and explained that again by the annihilation of the defects produced by irradiation.

The change in the UO_2 elastic modulus was studied as a function of different parameters; porosity concentration, stoichiometry, grain size, temperature, and dopant concentration [3, 4]. However limited data are available regarding the changes due to irradiation [3, 4]. Laux *et al.* studied the change of elastic modulus of nuclear fuel pellets and simulated spent fuel (SIMFUEL) with burn-up rate using acoustic microscopy [3]. The elastic modulus of SIMFUEL and irradiated fuel pellets decreased by about 25% as the burn-up increased up to 100 GWd/MTU. Beyond that, there is no data available for the irradiated fuel pellets but the elastic modulus of SIMFUEL stabilized [3]. On the other hand, Pujol *et al.* founded that the elastic modulus of simulated fuel increased with increasing the fuel burn-up [4]. Simulated fuel is UO_2 that has been mixed with various additives to simulate the compositional effects of the burn-up. Two different methods were used to calculate the elastic modulus; X-ray diffraction under high pressure and Knopp indentation test. Both measurements show that the elastic modulus increased with the burn-up. Calculated elastic constant values from indentation tests increase smoothly with burn-up, and saturate after burn-up of about 100 GWd/MTU, while the data obtained from the compressing test increase lineally with burn-up.

In general, there is limited amount of data available about the mechanical properties of UO_2 after irradiation [3 - 10], and some of them are contradicting especially regarding the changes in the elastic modulus [4]. Therefore, this work is intended to provide a systematic study of the change in UO_2

mechanical properties under heavy ion irradiation using different techniques and methods to study this phenomena; namely thin film geometry and nanoindentation.

Thin film geometry allows for control over several properties such as microstructure, oxygen stoichiometry, and impurity concentrations. This ability permits the advancement of the knowledge and understanding of the nanoscale behavior of uranium oxide under different irradiation and temperature conditions, which can lead to better burn-up performance. Nanoindentation has proved to be a useful technique to determine the mechanical properties for different non-nuclear materials [11 - 13] as well as nuclear materials such as; YSZ [14, 15], CeO_2 and UO_2 [16 - 18], simulated nuclear fuel [19], and irradiated materials [14, 15, 20, 21]. Nanoindentation is suitable for investigating the mechanical properties of thin films due to the ability to apply loads in the range of 10^{-9} N and limiting the depth of penetration to nanometers [20, 22]. In the current study, nanoindentation was used to study the change in mechanical properties of UO_2 thin films under different irradiation damage and temperature conditions. However, the substrate can affect the measurements [22]; therefore finite element modeling (FEM) was used to account for the substrate effect on the nanoindentation measurements [21]. In addition, FEM provides a method to estimate the mechanical properties of the irradiated UO_2 film layers independently from the unirradiated layers.

This work represents a systematic study regarding the change in the mechanical properties of UO_2 under heavy ion irradiation using single crystal thin films deposited on YSZ substrates. The irradiation conditions were chosen to assure that the radiation damage effect (displacement cascades and implanted gas bubbles) are confined within the UO_2 film, with no defects introduced into the substrate material. Finally, this study is focusing on using thin films of UO_2 as a surrogate for the bulk nuclear fuel that can be used to obtain similar and relevant results in better controlled conditions and without the dealing with large quantities of nuclear materials.

2. Experimental Methods

2.1 Film Deposition, Characterization, and Irradiation

Thin films of UO_2 were deposited on single crystal YSZ (8 %mole Y_2O_3) substrates using reactive gas magnetron sputtering technique [23, 24]. The sputtering was performed in current control mode, in which the ion current was fixed at 0.056 mA, with the power and voltage were maintained at approximately 20 W and 352 V respectively. Before the deposition process, the substrate was annealed at 400°C for two hours followed by two hours annealing at 750°C. During the deposition process, the temperature was fixed at 700°C and the oxygen partial pressure was 10^{-5} Pa. The substrate was rotated at

60 RPM to insure uniform heat distribution and growth rate (3.4 Å/sec). The final thickness of the films is 5000Å estimated by Rutherford backscattering spectroscopy (RBS). X-ray diffraction measurements (XRD) were performed using PANalytical / Philips X'pert MRD X-ray instrument using Cu K- α 1 source producing x-rays with wavelength of 1.54056Å. The measurements performed using high-resolution parallel beam configuration with hybrid mirror and 2-bounce monochromator and high-speed line detector. XRD measurements show that the film has a single (2 0 0) growth orientation as shown in Figure 1. The UO₂ thin film structural characterizations were explained in more details elsewhere [24].

The microstructure of as-grown and irradiated UO₂ thin films was investigated using JEOL 2010 with 2010 LaB₆ filament transmission electron microscope (TEM) which is optimized for diffraction contrast imaging and capable of sample tilt along two perpendicular axes. TEM specimens were prepared by mechanically ground the YSZ side to about 50 μ m thickness using diamond lapping films. Then, ion milling with 3–5 keV Ar ions was used to thin the specimens to 30-60 nm thickness. Cross sectional TEM images revealed that UO₂ films were deposited on the YSZ substrate in columnar morphology extended from the film-substrate interface to the film surface, as shown in Figure 2. This columnar structure is common in the thin film deposition and was noticed in several studies [25 - 27]. Plan view TEM image show that UO₂ film has several dislocation lines which formed due the misfit between the film and substrate lattices [25, 28, 29], as shown in Figure 3. Finally, Thin films were irradiated with 600 keV Kr⁺ ions at doses of 1E14, 5E14, 1E15, 1E16 and 1.5E16 ions/cm² at 25°C and 600°C using High Voltage Engineering Van de Graaff accelerator. Figure 4 shows the Kr⁺ concentrations and differential energy deposition estimated by SRIM calculations [30]. Kr⁺ ions had a maximum range of about 4000Å in UO₂ with peak concentration at 1629Å. Irradiation results at 600°C were compared to one sample labelled “annealed sample” that was exposed to the same thermal history as the sample irradiated with dose of 1E16 ions/cm². It was kept at 600°C for approximately 90 minutes.

2.2 Nanoindentation and Finite Element Modeling

Nanoindentation measurements were performed using Hysitron TI 950 TriboIndenter, which provides a precise control over the applied load and the indentation depth and can apply load over the range of 30 nN to 10 N. All the measurements were performed at load control mode in which the maximum applied load was fixed at 5000 μ N. Each sample was indented using Berkovick indenter.

The experiment was performed into two steps; the loading step, when the load is applied to push the indenter into the material at a loading rate of 1 mN/sec until it reaches the maximum specified load (5000 μ N). Then the indenter is drawn back to its original position during the unloading step. It is worth mentioning that, the source of errors in the nanoindentation measurements are mainly coming from

estimation of the contact area between the indenter and the material; and it is generally accepted to be about 5% or less [20]. For each measurement, the material is indented at different nine different positions and the average value is used to provide statistically acceptable data. In the same time, the standard deviation (shown with the data below) used to show the spread of the data and assure its consistency.

Finite element modeling was used to simulate the nanoindentation process using the ABAQUS finite element program [31]. The film had a thickness of 0.5 μm , a width of 3 μm , while the substrate had a thickness of 2.54 μm and a width of 3 μm . The film-substrate unit was divided into small elements with density increased in the high deformation area and decreased on the boundaries to save computational time. The smallest element size was 15x20 nm^2 and was located just under the indenter, with a total of 3016 elements, as shown in Figure 5. The lower bottom of the substrate was fixed with no displacement or rotational movement, while the nodes on the axisymmetric line in the film and substrate as well as the indenter were allowed to move in the vertical direction only. The interface connection between film and substrate was assumed perfect, with no sliding. No direct force acted on the substrate and all the plastic deformation was confined to the film as verified by FEM. Therefore, the substrate was modeled as an elastic medium with the same elastic properties from the modeling of YSZ substrate ($E=175 \text{ GPa}$, $\nu = 0.3$) [32].

The indenter was described as rigid body that could not be deformed during the simulation. This is a valid approximation since the indenter is made out of diamond that shows very small elastic response (elastic modulus equal to 1140 GPa); this approximation might not be valid if the material is very hard such as diamond-like carbon [20]. All materials were simulated using classical metal plasticity model in ABAQUS [31], in which the mass density, Poisson's ratio, elastic modulus, and yield strength of each material are required. Work hardening can be modeled by introducing the work hardening exponent, but since both YSZ and UO_2 are ceramics, it was not included into the simulation. The mass density and Poisson's ratio were fixed in all simulations and materials were assumed to be locally isotropic since the probing area of nanoindentation is very small.

The UO_2 film was modeled as an elastic-plastic material with density of 10.97 g/cm^3 and Poisson's ratio of 0.3. The simulation used arbitrary values for elastic modulus and yield strength which were allowed to change to achieve the best fit for the experimental curve. Results showed that the elastic modulus is lower than values found in the literature [16], which can be attributed to the difficulty in simulating the exact shape of the indenter tip, and the anisotropy of the elastic modulus [32]. On the other hand, the FEM estimation of yield strength is much larger than the literature value for bulk UO_2 [33]. These differences are expected in thin films, and were observed previously in Cu thin films deposited on Si [34]. The high strength of thin films compared to bulk samples is attributed to the small crystal size and high dislocation density in the thin films, as shown in Figure 3. The density was calculated to be

$1.38\text{E}14$ dislocations/ m^2 which is even higher than the values reported for irradiated fuel pellets up to burn up of 40 GWD/t [35]. In addition, the substrate constrains the dislocation motion in thin film geometry [36]. In order to estimate the film hardness, a separate simulation was performed on a bulk simulated sample of UO_2 . This simulation used the elastic modulus and yield strength values estimated from the best fit of experimental data.

In this study, $5000\text{\AA}$ UO_2 films were irradiated with 600 keV Kr^+ at doses ranging from $1\text{E}14$ to $1.5\text{E}16$ ions/ cm^2 . According to SRIM calculations [30], the Kr^+ ion concentration associated with the deposition energy was confined within $4000\text{\AA}$ of the film thickness, as shown in Figure 4. Three layers were used in the FEM simulation; the first layer extended from the film surface up to a depth of $4000\text{\AA}$. The unirradiated film represented the remaining portion of the film to the film-substrate interface, and the substrate was simulated as the third layer. The top two layers of the film were divided into small elements. The smallest element size just under the indenter tip was $15 \times 20 \text{ nm}^2$, giving a total number of elements in the irradiated and unirradiated film layers of 1541 and 335 elements respectively, while the total number of elements in the substrate was 1340.

3. Results

UO_2 thin films with thicknesses of $5000\text{\AA}$ were irradiated with 600 keV Kr^+ ions over a dose range of $1\text{E}14$ to $1.5\text{E}16$ ions/ cm^2 at 25°C and at 600°C , all irradiation parameters estimated from SRIM calculations [30] are summarized in Table 1. Nanoindentation was used to measure the change in mechanical properties and then results were fitted using FEM to account for the substrate effect. The results are summarized as follow:

3.1 Irradiation at 25°C

Figure 6 shows the load-displacement curves for the as-grown film as well as the irradiated films. For the same maximum applied load ($5000 \text{ }\mu\text{N}$), all irradiated films have a smaller penetration depth compared to the as-grown film, which implies that the material is hardening with irradiation. Figure 7 shows the FEM best fitting for unirradiated $5000\text{\AA}$ UO_2 thin film during loading and unloading stages and the simulation fits the experimental data well in both stages.

FEM results show that the elastic modulus decreased with initial irradiation, then it increased at dose of $5\text{E}14$ ions/ cm^2 and followed by slight reduction at dose of $1\text{E}15$ ions/ cm^2 , but the elastic modulus kept increasing afterward with irradiation dose, as shown in Figure 8. Overall, the initial irradiation led to reduction in the elastic modulus, however further irradiation led to the increase of the elastic modulus by approximately 6%. At the dose of $1\text{E}16$ ions/ cm^2 , the elastic modulus reached to a value of 175 GPa,

about 3% lower than the original value before irradiation. For yield strength and hardness, both properties increased with initial irradiation dose as shown in Figures 9 and 10 respectively. Yield strength increased substantially after irradiation at $5\text{E}14$ ions/cm² then decreased at $1\text{E}15$ ions/cm² and finally slowly increased with dose. Hardness followed the same trend as yield strength but with less dramatic changes in early stages of irradiation. Overall, the yield strength and hardness increased approximately by 17% and 13% with irradiation respectively. All elastic modulus, yield strength, and hardness results estimated by FEM are summarized in Table 2.

Plan-view TEM images for film irradiated with 600 keV Kr⁺ ions at a dose of $1\text{E}16$ ions/cm² at 25°C show round features, with size of 1.18 ± 0.1 nm that have an opposite contrast in over and under focus imaging conditions, are shown in Figure 11. This is an indication of the formation of bubbles or voids in the film due to the Kr⁺ ions irradiation [37]. The increase in the hardness and the yield strength occurs via the formation of gas bubbles and dislocation loops into the UO₂ matrix that increase with the irradiation dose. These bubbles and loops impede the dislocation motion and so harden the irradiated films [38 - 40].

Elastic modulus represents a measurement of the material resistance to displacement of adjacent atoms [41]. The change in elastic modulus is always related to the change in the interatomic bonding, mainly due to the presence of point defects. The change in elastic modulus with irradiation can be due to two effects. First, the presence of vacancies or interstitial defects will change the total number and strength of the interatomic bonds in the system. Second, the point defects induce lattice expansion and associated change in bond forces [42, 43]. The reduction of elastic modulus with point defects has been reported for fluorite-structure in pure and gadolinium-doped CeO₂ by measuring the change in elastic modulus with different oxygen partial pressure at 800°C using nanoindentation [43, 44]. Therefore the initial decrease in elastic modulus after irradiation with $1\text{E}14$ ions/cm² is caused by the introduction of point defects and dislocation loops which led to the formation of large number of dislocation in the irradiated film ($3.63\text{E}14$ dislocations/m²) as shown in Figure 11. If the dislocation density is high enough, newly formed point defects and displacement cascades are more likely to react with the existing defect clusters and erase them [38 - 40]. Therefore, further irradiation at higher irradiation doses led to the increase in the elastic modulus as shown in Figure 8.

3.2 Irradiation at 600°C

Figure 12 shows the load-displacement curves for UO₂ thin films irradiated with 600 keV Kr⁺ at 600°C in addition to the as-grown sample and one annealed sample. For the same maximum applied load (5000 µN), the annealed sample has a higher penetration depth than the reference sample. This implies that, the sample was softened with annealing due to the recovery of material defects. For the sample

irradiated with a dose of $1\text{E}14$ ions/cm², there was a slight decrease in hardness, but as the irradiation dose increased, the yield strength and hardness increased systematically with dose, as shown in Figures 9 and 10 and as summarized in Table 2.

In order to explain the material behavior under irradiation, TEM plan view images were used to investigate the microstructure evolution. Figures 14 and 15 show that nanocrystallites formed in UO₂ thin films irradiated with 600 keV Kr⁺ ions at a dose of $1\text{E}14$ and $1\text{E}16$ ions/cm² at 600°C with average size of 3 and 2 nm respectively. The nanocrystallites number density and volume fraction increased with irradiation dose as shown by the increase of the intensity of the diffraction rings. The diameter of the nanocrystallites diffraction rings are smaller than the distance between the (0 0 2) diffraction spots. This is attributed to the change in orientation of the nanocrystallites. The interplanar distance for the newly formed ring was found to be 3.131Å, very close to the interplanar distance for (1 1 1) crystallographic orientation with 3.148Å [45]. TEM images show that gas bubble was formed after irradiation with 600 keV Kr⁺ ions at a dose of $1\text{E}14$ ions/cm² at 600°C, as shown in Figure 16. The measured bubble density and volume fraction are less than bubbles formed after irradiation at 25°C, which is attributed to the recrystallization and the associated formation of nanocrystallites observed at the 600°C.

Figure 8 shows that the elastic modulus decreased by approximately 9% after annealing at 600°C. This reduction is due to the thermal expansion [46, 47] of the lattice and so the increase in the lattice parameter of the UO₂ thin film by annealing from 5.470 to 5.478Å as measured by XRD and shown in Figure 13. After the irradiation with $1\text{E}14$ ions/cm², the elastic modulus decreased compared to the annealed film due to the introduction of point defects [42, 48]. As the irradiation dose increased to $1.5\text{E}16$ ions/cm², the elastic modulus systematically increased due to the recrystallization which led to the annihilation of point defects and dislocation loops [42, 43, 49, 50], as shown in Figure 8 and summarized in Table 2.

4. Discussions

In general terms, ceramic materials are hard and deform in a brittle manner. However they can deform plastically if the stress and temperature levels are allowing dislocations to be mobile [51]. Several studies have focused on the deformation mechanisms in UO₂ at different conditions and temperatures. Keller et al., found that the movement of screw dislocations control the deformation of single crystal UO₂ during microhardness testing at 600°C [52]. Ashbee and Yust suggested that the plastic deformation of superstoichiometric UO₂ is a combination of UO₂ and U₄O₉ attached to the active slip systems [53]. In addition, Fossati *et al.*, applied molecular dynamics to simulate dislocation motion in UO₂ and found that the critical stress for dislocation glide is decreasing with temperature [54]. These studies confirmed that

plastic deformation exists in UO_2 and so applying Orowan mechanism [38, 55, 56] model is relevant to understand the hardening mechanisms in UO_2 .

Nanocrystallites, bubbles and precipitates, harden the material by acting as obstacles for dislocation motion [38, 55]. The critical resolved shear stress (CRSS) for Orowan hardening is given by [56 - 58]:

$$\tau = 0.9 \frac{\mu b \left[\ln \left(2 / b \left(1 / L_p + 4 / \pi d \right) \right) \right]^{1.5}}{2\pi(1-\nu)^{0.5} L_p \left[\ln \left(L_p / b \right) \right]^{0.5}} \quad (1)$$

$$L_p = d \left[(\pi / 6f)^{0.5} - (\pi / 4) \right] \quad (2)$$

due to the presence of an array of obstacles in the path of a line dislocation. In these equations, τ is the critical resolved shear stress, μ is the shear modulus, b is the Burgers vector, ν is the Poisson's ratio, L_p is the edge to edge spacing between randomly spaced obstacles, d is the bubble diameter, and f is the bubble volume fraction. The shear modulus of UO_2 was found to be 76.3 GPa [59] and the Burgers vector for (2 0 0) single orientation film was estimated to be 0.274 nm. The CRSS formed due to the formation bubbles and nanocrystallites was calculated and summarized in Tables 3 and 4.

Gas bubbles implanted in all films irradiated at 25°C, act as obstacles for the dislocation motion within the films. When an external stress is applied, it causes the dislocation lines to bow between the obstacles/bubbles until the dislocation segments join and annihilate each other. Orowan hardening can be used to estimate the critical resolved shear stress (CRSS) for an array of obstacles based on the assumption that these obstacles are uniformly distributed in the material [57, 58, 60]. The CRSS was found to increase with the obstacles volume fraction, which follows the increase in irradiation dose as summarized in Table 3. In the same time, the yield strength was found to increase with the dose too as summarized in Table 2. The yield strength was plotted against the CRSS and found to increase linearly with slope of 1.6 as shown in Figure 17. The intercept represents the yield strength of the unirradiated UO_2 films (at CRSS = 0) [58] and the value of 8.7 GPa is about 16% higher than the values estimated by FEM as summarized in Table 2. This difference may be due to the poor fitting because of limited number of data points.

Irradiation of the films at 600°C led to a systematic increase in hardness with dose. Initially, the film hardness and yield strength decreased with annealing, as well as after the initial irradiation. Further irradiation increased the hardness and yield strength systematically with dose. TEM showed the formation of nanocrystallites associated with the irradiation at 600°C which is a characteristic signature of recrystallization attributed to the release of internal energy of the system [49]. The nanocrystallite act as obstacles for the motion of dislocations and induce hardening. Orowan hardening was employed to estimate CRSS as summarized in Table 4. Gas bubbles were observed with TEM images and had an

approximate diameter of 0.7 nanometers. These bubbles are smaller than those formed during irradiation at 25°C. It is believed that recrystallization led to the loss of some of the implanted gases [50]. It is clear that the induced strength from the nanocrystallites is much larger than the strength induced by the bubbles. This implies that, the hardening of irradiated UO_2 films at 600°C is primarily driven by the formation of the nanocrystallites in the film.

It is worth mentioning that, the mechanism of Orowan hardening is based on the assumption that obstacles are uniformly distributed throughout the material [38, 55, 56]. This assumption is acceptable for bubbles formed in the irradiated films due to the spread of bubble positions. On the other hand, nanocrystallites with different orientations can be connected together in clusters as shown in Figures 14 and 15. This is one point in which the mechanism of Orowan hardening may not be able to describe accurately due to the underlying complexity of the microstructure. The yield strength of UO_2 films irradiated at 600°C is plotted against the CRSS induced by gas bubbles and nanocrystallites, as shown in Figure 18. The yield strength increased linearly with the CRSS, which followed the Orowan hardening mechanism. The slope of the line of best fit is known as Schmid factor [61] or Taylor factor [57, 58]. The Schmid factor from Figure 17 is 1.4, less than the value 1.8, obtained from He-bubbles in Ni [58] and Al precipitates in Ni [60]. This difference can be attributed to the loss of some implanted gases due to the recrystallization, internal stresses in the thin films, and the differences between the metallic Ni and the ceramic UO_2 .

Hardness is the material resistance to plastic deformation. Empirical observations led to a linear relationship between the hardness and yield strength in metals [62]. From this relation, the hardness of a ductile metal is found to be approximately three times its yield strength [63 - 65]. Thin films are known to be harder than their bulk sample counterparts [56, 66]. Thus, the relationship between hardness and yield strength of thin films is expected to deviate; especially for ceramic materials. The hardness and yield strength of UO_2 thin films irradiated with 600 keV Kr^+ ions at 25°C and 600°C were found to increase with dose as summarized in Table 2. The hardness plotted against the yield strength values estimated by FEM as shown in Figure 19. When a linear fit was used, the resulting slope was 1.54. These results can be used as a base to estimate the mechanical properties of thin films using the nanoindentation measurements.

Finally, these results show that elastic modulus initially decreased with irradiation at 25°C and 600°C, but it increased with higher irradiation doses at both temperatures similar to Pujol *et al.* results [4]. In addition, the study showed that irradiation can lead to recrystallization of the nuclear fuel which also has been reported in other studies [7, 8]. Therefore, this work verified that UO_2 thin films can be used to obtain similar and relevant results for bulk fuel samples to understand the nuclear fuel performance in controlled experimental conditions [67 - 70] and using small scale mechanical testing [71, 72].

5. Conclusions

This study focused on the change in mechanical properties of single crystal UO_2 thin films under different irradiation conditions. The mechanical response of thin films of UO_2 due to radiation damage and the formation of gas bubbles was investigated under irradiation with 600 keV Kr^+ ions at two different temperatures, 25°C and 600°C. At 25°C; the film hardness and yield strength were found to increase with irradiation dose due to the formation of gas bubble formation and dislocation loops. On the other hand, the elastic modulus was found to decrease at the initial irradiation dose, and then it increased with increasing the irradiation dose. When irradiation performed at 600°C, the annealing effect was noticed mainly as a decrease in the elastic modulus due to the annihilation of point defects in the film. Both hardness and elastic modulus decreased at the lowest irradiation dose, and then they increased systematically with dose. This behavior is attributed to the formation of nanocrystallite with diameter and densities increase with dose. Although gas bubbles were formed after irradiation, but nanocrystallites represent the primary source of hardening. The film hardness was found to increase with dose and follow Orowan hardening mechanism. Finally, by comparing these results to the previous studies focusing on the mechanical properties of nuclear fuel, it was concluded that thin films can be used to get results that are similar and relevant to bulk fuel samples.

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Table 1. Irradiation parameters for Kr⁺ ions.

Ion	Energy (keV)	Range (Å)	Dose (ions/cm ²)	Average atomic concentration (at. %)	Average dpa
Kr ⁺	600	1629	1E14	0.02	<1
			5E14	0.10	4
			1E15	0.2	8
			5E15	1.01	43
			1E16	2.03	86
			1.5E16	3.04	130

Table 2. FEM estimation of the elastic modulus, yield strength, and hardness of untreated, annealed, and irradiated 5000Å UO₂ thin films irradiated with 600 keV Kr⁺ ions at 25°C and 600°C.

Dose (ions/cm ²)	Temperature (°C)	Elastic modulus (GPa)	Yield strength (GPa)	Hardness (GPa)
Reference	25	180	7.5	13.5
1E14	25	165	8.5	14.2
5E14	25	170	9.7	14.9
1E15	25	167	9.5	14.5
5E15	25	170	9.7	14.9
1E16	25	175	9.8	15.3
Annealed	600	160	7.2	12.5
1E14	600	145	7.0	11.6
1E15	600	155	7.5	12.5
5E15	600	165	8.2	13.6
1E16	600	175	9.2	14.8
1.5E16	600	178	10.0	15.5

Table 3. Bubbles size, density, and the critical resolved shear stress for UO₂ films irradiated with 600 keV Kr⁺ ions at 25°C.

Dose	Bubble diameter (nm)	Bubble density (nm ⁻³)	CRSS (GPa)
1E14	1.18±0.1	5.8E-6	0.2652
5E14	1.18±0.1	9.6E-6	0.2835
1E15	1.18±0.1	3.4E-5	0.3021
5E15	1.18±0.1	8.3E-5	0.5326
1E16	1.18±0.1	1.4E-4	0.6928

Table 4. Nanocrystallites and bubbles sizes, densities, and the critical resolved shear stress for UO₂ films irradiated with 600 keV Kr⁺ ions at 600°C with different irradiation doses.

Dose	Nanocrystallites			Bubbles		
	diameter (nm)	density (nm ⁻³)	CRSS (GPa)	diameter (nm)	density (nm ⁻³)	CRSS (GPa)
1E14	3 ±0.5	1.8E-4	3.865	0.7 ±0.07	7.3E-5	0.2116
1E15	1.4±0.5	3.4E-4	4.734	0.7 ±0.07	4.0E-5	0.142
5E15	1.7±0.5	4.3E-4	5.314	0.7 ±0.07	2.1E-5	0.105
1E16	2 ±0.5	5.4E-4	5.730	0.7 ±0.07	8.6E-6	0.0644
1.5E16	2±0.5	5.5E-4	5.812	0.7 ±0.07	7.9E-6	0.0621

Figure 1. Indexed specular $2\theta - \Omega$ scan for 5000Å UO_2 film deposited on YSZ substrate.

Figure 2. Bright-field cross sectional TEM image for sample 49. The image shows the columnar grains of the UO_2 thin film above the substrate.

Figure 3. Bright-field, under focus and plan view TEM image for as-grown UO_2 thin film. Black arrows are pointing to dislocation lines.

Figure 4. Kr^+ concentrations (dark line) and differential energy deposition (dotted line) in 5000 Å UO_2 thin film irradiated with 600 keV Kr^+ ions at dose of $1\text{E}14$ ions/ cm^2 .

Figure 5. Elements distribution throughout the film-substrate unit.

Figure 6. Load – displacement curve for UO_2 thin films deposited on YSZ substrate and irradiated with 600 keV Kr^+ ions at 25°C with different irradiation doses.

Figure 7. Best FEM fitting for load – displacement curve for 5000Å UO_2 thin films deposited on YSZ substrate.

Figure 8. FEM estimation of the elastic modulus versus dose for UO_2 thin films irradiated with 600 keV Kr^+ ions at both 25°C and 600°C. E_0 and E_T are the elastic modulus of the untreated and the annealed samples respectively. The error bars will represent the standard deviation in the nanoindentation elastic modulus measurements.

Figure 9. FEM estimation of the yield strength versus dose for UO_2 films irradiated with 600 keV Kr^+ ions at both 25°C and 600°C. σ_0 and σ_T are the yield strength of the untreated and the annealed samples respectively. The error bars will represent the standard deviation in the nanoindentation hardness measurements.

Figure 10. FEM estimation of the hardness versus irradiation dose for UO_2 films irradiated with 600 keV Kr^+ ions at both 25°C and 600°C. H_0 and H_T are the hardness of the untreated and the annealed samples respectively. The error bars will represent the standard deviation in the nanoindentation hardness measurements.

Figure 11. Plane view TEM images for UO_2 thin film irradiated with 600 keV Kr^+ ions at a dose of $1\text{E}16$ ions/ cm^2 at 25°C . Under and over focused conditions showed the voids that are surrounded by dotted white circles for visualization.

Figure 12. Load – displacement curve for UO₂ thin films deposited on YSZ substrate and irradiated with 600 keV Kr⁺ ions at 600°C with different irradiation doses.

Figure 13. The development of the (200) UO_2 reflection for as-grown, annealed at 600°C, irradiated with 600 keV Kr^+ ions at 600°C with 1E14 and 1E16 ions/cm². The dotted line shows the original peak position of the as-grown film.

Figure 14. Plan view image for UO₂ film irradiated with 600 keV Kr⁺ ions at 600°C with 1E14 ions/cm².
The arrows pointed to different nanocrystallites.

Figure 15. Plan view image for UO_2 film irradiated with 600 keV Kr^+ ions at 600°C with $1\text{E}16$ ions/ cm^2 .

The arrows pointed to different nanocrystallites.

Figure 16. Plane view TEM images for UO_2 thin film irradiated with 600 keV Kr^+ ions at a dose of $1\text{E}14$ ions/ cm^2 at 600°C . Under and over focused conditions showed the voids that are surrounded by dotted white circles for visualization.

Figure 17. Yield strength of 5000Å UO₂ thin films versus CRSS evaluated using Orowan hardening mechanism due to the presence of bubbles after irradiation with 600 keV Kr⁺ ions at 25°C.

Figure 18. Yield strength of 5000Å UO₂ thin films versus CRSS evaluated using Orowan hardening mechanism due to the presence of nanocrystallites after irradiation with 600 keV Kr⁺ ions at 600°C.

Figure 19. Hardness versus yield strength for UO_2 films irradiated with 600 keV Kr^+ ions at 600°C.