Epitaxial Fe/Y₂O₃ interfaces as a model system for oxide-dispersion-strengthened ferritic alloys

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The fundamental mechanisms underlying the superior radiation tolerance properties of oxide-dispersion-strengthened ferritic steels and nanostructured ferritic alloys are poorly understood. Thin film heterostructures of Fe/Y₂O₃ can serve as a model system for fundamental studies of radiation damage. Epitaxial thin films of Y₂O₃ were deposited by pulsed laser deposition on 8% Y:ZrO₂ (YSZ) substrates with (100), (110), and (111) orientation. Metallic Fe was subsequently deposited by molecular beam epitaxy. Characterization by x-ray diffraction and Rutherford backscattering spectrometry in the channeling geometry revealed a degree of epitaxial or axiotaxial **orientation** for Fe(211) deposited on Y₂O₃(110)/YSZ(110). In contrast, Fe on Y₂O₃(111)/YSZ(111) was fully polycrystalline, and Fe on Y₂O₃(100)/YSZ(100) exhibited out-of-plane texture in the [110] direction with little or no preferential in-plane orientation. Scanning transmission electron microscopy imaging of Fe(211)/Y₂O₃(110)/YSZ(110) revealed a strongly islanded morphology for the Fe film, with no epitaxial grains visible in the cross-sectional sample. Well-ordered Fe grains with no orientation to the underlying Y₂O₃ were observed. Well-ordered crystallites of Fe with both epitaxial and non-epitaxial orientations on Y₂O₃ are a promising model system for fundamental studies of radiation damage phenomena. This is illustrated with preliminary results of He bubble formation following implantation with a helium ion microscope. He bubble formation is shown to preferentially occur at the Fe/Y₂O₃ interface.

1. Introduction

Next-generation fission and future fusion reactors require the development of highly radiation-damage-resistant materials which can be easily fabricated into large-scale components. Dramatic increases in radiation tolerance, along with improved high temperature mechanical properties, can likely be achieved by introducing nanoscale complexity such as interfaces into bulk materials [1]. Nanoscale interfaces and other defects can act as effective sinks for radiation damage, without the mechanical drawbacks typically associated with microscale interfaces and grain boundaries [2]. Oxide dispersion strengthened (ODS) ferritic steels and nanostructured ferritic alloys (NFAs), consisting of nanoscale, complex Y-Ti-O oxide precipitates in a ferritic steel matrix, have been extensively investigated as a promising route to achieve superior radiation, temperature, and stress tolerance [2]. These materials have been shown to achieve high tensile, creep, and fatigue strength, thermal stability, and neutron radiation resistance [3]. Nanoscale oxide particles promote radiation-induced defect recombination and trap transmutation-produced He in small high pressure bubbles, as well as increase high temperature strength and creep resistance by impeding dislocation climb and glide [3]. Although ODS ferritic steels and NFAs have been studied in detail with state-of-the-art materials characterization techniques [3-7] and theoretical simulations based on density functional theory (DFT) [8], the detailed physical mechanisms behind these remarkable properties are still poorly understood.

Recently, we have explored the effect of nanoscale defects with epitaxial thin films as a model system; the density of nanoscale misfit dislocations can be controlled by varying the film lattice parameter via alloying [9, 10]. Here, we employ a similar method to explore the fundamentals of radiation damage resistance in ODS ferritic steels and nanostructured ferritic alloys by attempting to synthesize epitaxial interfaces of Fe/Y₂O₃. Not only would this provide a convenient, well-controlled model system to study interfacial radiation damage behavior over a much larger spatial area than that offered by nano-scale precipitates, but coherent and semi-coherent interfaces have been observed between nano-scale Y-Ti-O precipitates and the Fe

matrix in both ODS ferritic steels [7] and NFAs [3], indicating that the properties of coherent interfaces may be directly applicable to real-world materials. To this end, the structural characterization of epitaxial Y_2O_3 on $\text{Y}:\text{ZrO}_2$ (YSZ) substrates, and subsequent Fe deposition, is presented with detailed information on the epitaxial orientation and interface structure of the Fe/ Y_2O_3 heterostructure. Preliminary results of He ion implantation utilizing a helium ion microscope in conjunction with scanning transmission electron microscopy (STEM) [11] indicate preferential He bubble formation at the Fe/ Y_2O_3 interface.

2. Experimental Methods

Epitaxial thin films of Y_2O_3 were deposited on 8% $\text{Y}:\text{ZrO}_2$ (YSZ) substrates with (100), (110), and (111) orientations by pulsed laser deposition [12] from a pure Y_2O_3 target. A KrF laser operating at 248 nm, 4 Hz, and a fluence of $\sim 2.4 \times 10^4 \text{ J/m}^2$ on the target was utilized for laser ablation. The substrate temperature was held at 750°C in a background pressure of 1.33 Pa O_2 . Y_2O_3 films were deposited to a thickness of 200 nm. After *ex situ* structural characterization by x-ray diffraction (XRD), the samples were loaded into an ultra-high vacuum (UHV) molecular beam epitaxy (MBE) system [9, 10]. Samples were cleaned of adventitious carbon contamination by exposure to activated oxygen generated in an electron cyclotron resonance microwave plasma source at room temperature for 30 minutes. Samples were heated to 575-650°C for deposition of Fe from an effusion cell (flux rate 0.012 nm/s) to a nominal thickness of 50 nm. Reflection high energy electron diffraction (RHEED) patterns were monitored to observe the crystalline order of the cleaned Y_2O_3 surface and the subsequent Fe deposition. In some cases, *in situ* x-ray photoelectron spectroscopy (XPS, not shown) was utilized to confirm the removal of C, and lack of other impurities, on the Y_2O_3 surface after plasma exposure, and the presence of metallic Fe after deposition.

High resolution x-ray diffraction (XRD) patterns were collected on a PANalytical X'Pert Materials Research Diffractometer (MRD) using $\text{Cu K}\alpha_1$ radiation monochromated with a hybrid mirror/4 crystal monochromator, resulting in an incident beam divergence of 0.0033° (12 arcsec). Low resolution XRD patterns were collected on a PANalytical X'Pert MultiPurpose

Diffraction (MPD) with an incident beam divergence of 0.05° provided by a curved x-ray mirror. Pole figures to determine in-plane orientations were collected on a Rigaku MicroMax-007 HF diffractometer paired with an imaging plate detector. Pole figures were reconstructed from large-area diffraction patterns collected at 5° steps in ϕ . Rutherford backscattering spectrometry (RBS) measurements with 2 MeV He^+ ions (National Electrostatics Corporation 9SDH-2 tandem electrostatic accelerator) in the channeling geometry were utilized to measure crystalline quality and atomic order in the films. Electron backscatter diffraction (EBSD) was performed using a JEOL7600F field emission scanning electron microscope (FESEM) equipped with a Nordlys camera and interfaced with the Oxford/HKL Channel 5 software package. EBSD mapping was performed using an accelerating voltage of 20kV and step size of 20nm. The lattice parameter for bcc iron used for indexing was 0.2866 nm. Post-acquisition data processing was performed by first removing wild spikes, then smoothing to 6 nearest neighbors. As a final step, a single pass Kuwahara filter using a 3x3 grid and a 5° smoothing angle was applied. Pole figures were generated from the complete data set as upper hemisphere stereographic projections.

High resolution transmission electron microscopy (HRTEM) and STEM images were collected on a Titan 80-300 S/TEM operated at 300 kV. The microscope is fitted with a spherical aberration corrector for the probe-forming lens. The STEM high-angle annular dark-field images (STEM-HAADF) were collected with a collection angle ranging from 52 to 200 mrad. The electron probe diameter is approximately 0.1 nm with a probe convergence angle of 17.8 mrad. The cross-sectional TEM samples were prepared by using focused ion beam (FIB) sectioning in a dual beam SEM. He ions were implanted into a thin cross sectional TEM sample (180 – 300 nm thick, as determined in TEM by tilting the sample and measuring the separation of features of both the bottom and top surfaces) at room temperature using a helium ion microscope (Carl Zeiss, Germany) with a beam energy of 25 keV. The He^+ beam was scanned across an area of several square microns, which included Fe, Y_2O_3 , and their interface, with a fluence of $10^{22}/\text{m}^2$ [11]. Microstructure of the sample before and after the He ion implantation were analyzed in cross-section using HRTEM, electron diffraction, and HAADF-STEM.

3. Results and Discussion

3.1 Y_2O_3 on YSZ

When 9.5% Y is added to ZrO_2 , the crystal is stabilized in the cubic calcium fluorite structure with $a = 0.5125 \text{ nm}$. Y_2O_3 crystallizes in the cubic bixbyite structure with $a = 1.0604 \text{ nm}$ [13]. Because the bixbyite structure can be considered a defected fluorite structure with a doubled lattice parameter [14], epitaxy is possible on cubic fluorite substrates such as YSZ. The cube-on-cube lattice mismatch between the YSZ substrate and the Y_2O_3 film is calculated as $\delta = (a_{Y_2O_3} - 2 \cdot a_{YSZ}) / (2 \cdot a_{YSZ}) = 3.5\%$. The inverse system, YSZ on Y_2O_3 , has been successfully deposited by PLD with an epitaxial relationship [15]. Additionally, bixbyite In_2O_3 and Sn-doped In_2O_3 (ITO) have been deposited on YSZ as epitaxial layers utilizing oxygen-plasma-assisted molecular beam epitaxy [14, 16] and pulsed laser deposition [17, 18].

Figure 1 plots the high resolution θ - 2θ XRD patterns for Y_2O_3 films deposited on YSZ(100), YSZ(110), and YSZ(111) substrates. In all cases, epitaxial films with the same crystalline orientation as the substrate were obtained. Surprisingly, even at this thickness, the Y_2O_3 films have not fully relaxed to the bulk lattice parameters. Instead, the out-of-plane d -spacings observed in Fig. 1 correspond to an expansion of 0.6 – 0.9% relative to the bulk values of Y_2O_3 . For Y_2O_3 (100), this corresponds to an out-of-plane lattice parameter of 1.0668 nm . This expansion is consistent with a slight in-plane contraction of the Y_2O_3 film to better match the smaller lattice parameter of the YSZ substrate. The contraction of the in-plane lattice parameter to match the YSZ substrate can be observed clearly in the reciprocal space maps (RSMs) shown in the insets to Fig. 1(b) and Fig. 1(c) for Y_2O_3 /YSZ(110) and Y_2O_3 /YSZ(111), respectively. In both cases, substantial diffraction intensity is observed at the same q_{xy} value as the substrate diffraction peak, indicating the film and substrate possess the same in-plane lattice parameter. However, the film diffraction peaks are broad in the q_{xy} direction, indicating partial relaxation in some regions of the film.

No secondary orientations were observed for Y_2O_3 films deposited on YSZ(100) and YSZ(111), but for the film on YSZ(110) a small peak appears at $2\theta \approx 49.1^\circ$ ($d \approx 0.185 \text{ nm}$). The origin of this peak is unclear; the d -spacing is not consistent with a more highly strained region with an in-plane lattice parameter closer to the YSZ(110) substrate, since the expected Y_2O_3 (440) d spacing would be $> 0.1874 \text{ nm}$. Alternatively, the Y_2O_3 (433) reflection appears at a similar position ($2\theta = 50.129^\circ$, $d = 0.18183 \text{ nm}$), but an expansion of 1.9% would be required to shift the peak to the position observed in Fig. 1(b).

Out-of-plane rocking curves for the three film/substrate orientations are presented in Fig. 2. The YSZ substrates exhibit narrow rocking curve widths in the range of $0.005 - 0.02^\circ$ (full width at half maximum, FWHM). The rocking curve widths of the Y_2O_3 films are substantially broader than those of the substrates, with FWHM values in the range of $0.6 - 1.3^\circ$. This is consistent with the broad diffraction peaks observed in the RSMs. Such broad rocking curve widths indicate that, although single crystal epitaxy was achieved, the films possess a substantial defect structure. In addition, a lattice tilt of approximately 0.4° is observed for Y_2O_3 / YSZ(111). Lattice tilt has been observed for bixbyite In_2O_3 islands deposited on YSZ, but the tilt is only observed on YSZ(100) substrates [14, 19]. However, the lattice parameter of In_2O_3 is smaller than that of YSZ, such that epitaxial In_2O_3 experiences tensile strain on YSZ. In contrast, Y_2O_3 has a larger lattice parameter than YSZ and thus epitaxial films are under compressive strain. Epitaxial strain can drive lattice tilt to better match the substrate [19]; with a film of larger lattice parameter, tilt may be favored on YSZ(111) over (100) and (110).

3.2 Fe on Y_2O_3 /YSZ

After deposition by PLD and *ex situ* characterization by XRD, the Y_2O_3 /YSZ samples were loaded individually into the MBE chamber for Fe deposition. Figure 3(a) shows the RHEED patterns of the Y_2O_3 (100) film surface before Fe deposition. A well-defined streaky pattern indicates the film surface is reasonably flat and well-ordered, with a 2x reconstruction apparent along the Y_2O_3 [110] direction. After deposition of 50 nm Fe, the RHEED pattern

becomes spottier, indicating a roughening of the surface. The dominant in-plane spacing of the RHEED images does not change significantly along the two electron beam incidence directions, which implies that Fe does not show strong in-plane orientation; faint rings due to crystal grains randomly oriented in-plane are also visible in the RHEED images.

Figure 3(b) shows the low resolution XRD pattern for Fe / Y_2O_3 / YSZ(100). Under these measurement conditions, the $\text{Y}_2\text{O}_3(200)$ and $\text{Y}_2\text{O}_3(600)$ reflections are visible. In addition, a peak corresponding to Fe(110) is present. The absence of Fe peaks corresponding to other crystalline orientations means that Fe is oriented out-of-plane with the (110) plane parallel to the film surface. However, since the (110) surface is the close-packed surface for the bcc crystal structure, and thus typically favored as the growth surface even if no in-plane orientation is present, further information is required to determine the presence of epitaxial matching to Y_2O_3 . Figure 3(c) plots the Fe{110} pole figure for the same film. A diffraction ring is observed at a polar angle of 30° , corresponding to diffraction from Fe(101) planes which make an angle of 60° with the Fe(110) surface. The Fe(110) film exhibits weak in-plane texture with a 45° rotation relative to the underlying Y_2O_3 substrate, such that $\text{Fe}[001] \parallel \text{Y}_2\text{O}_3[110]$. However, the significant diffraction intensity at all other azimuthal angles to form a ring-like diffraction pattern indicates that the film exhibits many crystallites which are oriented out-of-plane but exhibit random in-plane orientations.

Comparable RHEED images for Fe / Y_2O_3 / YSZ(110) are shown in Fig. 4(a). Before Fe deposition, the $\text{Y}_2\text{O}_3(110)$ film surface displays a streaky RHEED pattern, again indicating a smooth, well-ordered surface. The $\text{Y}_2\text{O}_3(110)$ surface exhibits distorted tetragonal symmetry; although symmetric RHEED patterns were obtained every $25 - 30^\circ$ in azimuth, indexing them to the crystalline directions is not straightforward. Fe deposition leads to a very sparse, spotty pattern which nonetheless changes significantly when the azimuthal angle is changed by 26° . The XRD pattern plotted in Fig. 4(b) indicates that the Fe film is predominantly oriented in the Fe(211) direction, although a small contribution from Fe(110) is also visible. The Fe{110} pole figure in Fig. 4(c) confirms a strong epitaxial alignment with the underlying $\text{Y}_2\text{O}_3/\text{YSZ}(110)$, with two sets of intense diffraction peaks rotated azimuthally 90° from each other. The set appearing at a polar angle of $\sim 60^\circ$ arise from Fe(101) and Fe(110) planes, which make an angle of 30°

with Fe(211). In contrast, the orthogonal Fe{110} planes, Fe(011) and Fe($10\bar{1}$), are not equivalent with respect to Fe(211). The Fe(011) plane makes an angle of $\sim 54.7^\circ$ with Fe(211), placing it at $\sim 35^\circ$ on the pole figure, while the Fe($10\bar{1}$) plane makes an angle of $\sim 73.2^\circ$ with Fe(211), placing it out of range on the pole figure (it would appear at $\sim 16.8^\circ$). The presence of two diffraction peaks separated by 180° in azimuth, both arising from Fe(011) planes, indicates that the Fe(211) film possesses two domains rotated by 180° . A similar two-domain structure has been observed previously for Cr(211) films deposited on MgO(110) [20].

EBSD was utilized in an effort to further illustrate the orientation relationships identified from XRD for Fe / Y_2O_3 / YSZ(110). The EBSD inverse pole figure map (Fig. 5, top) shows a uniform grain size distribution of the Fe film in the submicron range. Pole figures generated from the map (Fig. 5, bottom) indicate the film has multiple texture components. The pole figures also depict very distinct and symmetrical arcs. Portions of similar arcs can be observed in the XRD pole figure (Fig. 4(c)). Pole figures of this form are typically a result of thin film axiotaxy. Although the crystal structures for Fe and Y_2O_3 are very different, certain planes in the substrate and film exist which share very similar d -spacing. Alignment of these planes during film growth manifests as a fiber texture lying off-normal to the sample surface [21]. The result is an epitaxial film which has an interfacial structure exhibiting periodicity along a single direction [22]. For this specimen, the pole figure analysis shows the simultaneous occurrence of different texture components within the same film, consistent with the XRD data. The {211} pole figure shows a partial clustering of poles in the film growth direction, indicating that some grains exhibit a preferred epitaxial orientation of Fe(211) $\parallel$ Y_2O_3 (110).

In contrast to the films discussed above, the surface of Y_2O_3 / YSZ(111) is extremely rough, as deduced from the spotty RHEED images shown in Fig. 6(a). As might be expected, Fe deposition on this surface proceeded poorly, resulting in a RHEED pattern which consists entirely of diffraction rings. From this pattern, the Fe film appears to be fully polycrystalline with no preferential in-plane orientation. The polycrystalline nature of the Fe film is confirmed by XRD in Fig. 6(b), which reveals Fe(110), Fe(200), and Fe(211) peaks with approximately the intensity distribution expected for a polycrystalline sample of Fe. The origin of the peak

appearing at $\sim 15.1^\circ$ (marked by an asterisk in Fig. 6(b)) is unknown; it cannot be indexed to Fe, Y_2O_3 , or YSZ. The pole figure of a polycrystalline material consists of uniform diffraction intensity at all polar and azimuthal angles. The pole figure in Fig. 6(c) exhibits some slight structure, but the overall intensity is very low and it is most likely that the structure arises from diffraction intensity spillover of Y_2O_3 or YSZ peaks.

A complementary measure of the degree of Fe epitaxy on Y_2O_3 /YSZ is to compare RBS backscattering in the random and channeling (normal to the film surface) geometries. Figure 7 plots the backscattering yield in both geometries for Fe films deposited on Y_2O_3 /YSZ(100), Y_2O_3 /YSZ(110), and Y_2O_3 /YSZ(111), respectively. In all three cases, the thickness of the Y_2O_3 layer (approximately 200 nm) places the Zr edge of YSZ near or under the Fe film peak. However, the backscattering yields of Y and Zr are very similar, so only a small jump in the backscattering signal occurs at the Zr edge. In the inset to Fig. 7(a), only slight channeling of the Fe film signal is observed, although the Y_2O_3 and YSZ(100) layers exhibit a reasonable decrease in intensity in the channeling geometry. The weak epitaxy observed by XRD (Fig. 3) does not result in a well-ordered crystalline structure perpendicular to the film surface. In contrast, the Fe film deposited on Y_2O_3 /YSZ(110) exhibits stronger channeling, with a minimum yield of $\chi_{\min} = I_{\text{random}}/I_{\text{channeling}} \sim 44\%$. As expected, the polycrystalline Fe film deposited on Y_2O_3 /YSZ(111) exhibits no channeling in Fig. 7(c).

From the XRD data in Fig. 4 and RBS channeling data in Fig. 7, it is clear that Fe exhibits no epitaxial orientation relative to Y_2O_3 /YSZ(111). This is in contrast to experimental TEM studies [7] and DFT simulations [8] of the Fe/ Y_2O_3 interface of an ODS ferritic alloy, which found the Fe(110) $\parallel$ Y_2O_3 (111) interface was stable, as well as a sink for point defects. The Fe(110) $\parallel$ Y_2O_3 /YSZ(111) epitaxial orientation may not have occurred in this work due to the rough surface morphology of the Y_2O_3 (111) surface. Alternatively, in both the ODS ferritic alloy and in the DFT simulation, both the Fe and the Y_2O_3 could experience strain to accommodate the lattice mismatch at the interface. However, when Fe is deposited on a thick film of Y_2O_3 , as occurred in this work, only the Fe is likely to experience epitaxial strain to accommodate the interface; the Y_2O_3 is immobilized due to its much greater thickness and mechanical clamping to

the YSZ substrate. The epitaxial orientation may not be favorable when the strain is entirely accommodated by the Fe film.

In contrast, it appears that Fe(211) deposits with reasonable epitaxial quality in some regions on $Y_2O_3(110)$ / YSZ(110). The out-of-plane orientation is clearly Fe(211) $\parallel$ $Y_2O_3(110)$. To determine the in-plane orientation from the pole figure in Fig. 4, it is necessary to collect a pole figure for Y_2O_3 or YSZ with the sample in the same orientation. That pole figure (not shown) produced a YSZ(004) reflection at $\sim 270^\circ$. Thus, the Fe[111] direction and YSZ[004] direction occur at the same azimuthal angle relative to the sample surface. This is consistent with an in-plane epitaxial orientation that can be described as Fe($\bar{1}11$) $\parallel$ $Y_2O_3(001)$.

To understand the atomic arrangement and lattice mismatch at the Fe(211)/ $Y_2O_3(110)$ interface, the $Y_2O_3(110)$ surface must first be elucidated. As discussed above, Y_2O_3 crystallizes in the cubic bixbyite structure (space group $Ia\bar{3}$). The $Y_2O_3(110)$ surface does not present a perfectly flat layer of atoms, as illustrated in side view in Fig. 8(a). In contrast, the Fe(211) surface is flat and reasonably close-packed in the bcc crystal system, as seen in Fig. 8(b). The arrangement of atoms at the Fe(211)/ $Y_2O_3(110)$ interface is shown in Fig. 8(c). Because it provides the best lattice match to Fe, only the Y cations from a single, buckled layer of Y_2O_3 are shown below the Fe(211) atom plane. A reasonable lattice match is achieved. The lattice spacing of Fe in the Fe[$0\bar{1}1$] direction is 0.4055 nm; the Y spacing alternates between two spacings of 0.4001 nm and two spacings of 0.3514 nm in the Y_2O_3 [$\bar{1}10$] direction. The lattice mismatch with Fe alternates between $(d_{\text{film}} - d_{\text{substrate}})/d_{\text{substrate}} \times 100\% = 1.34\%$ and 15.4%, respectively, with an average mismatch of 7.9%. This corresponds to an axiotaxial alignment of the Fe{100} planes with the Y_2O_3 {100} planes. In the perpendicular direction the situation is similar, although in this case the Y spacing along the [001] direction alternates between 0.5303 nm and 0.5348 nm. The Fe spacing along the [$\bar{1}11$] direction is only 0.2483 nm, approximately half the Y spacing. The lattice mismatch is then $(d_{\text{film} \times 2} - d_{\text{substrate}})/d_{\text{substrate}} \times 100\% = -6.4\%$ and -7.1%, respectively. The fairly large lattice mismatches estimated for the the Fe(211)/ $Y_2O_3(110)$ interface help explain why epitaxy wasn't strongly favored during the deposition, and

other orientations of Fe also nucleated. Nonetheless, optimization of the deposition parameters may produce films with an increased fraction of epitaxial orientation.

3.3 Microstructure and He⁺ implantation of Fe(211) on Y₂O₃/YSZ(110)

To better understand the epitaxy of Fe(211) on Y₂O₃/YSZ(110), the microstructure was investigated by high resolution TEM and STEM. Figure 9(a) shows a bright field (BF) TEM micrograph of Fe/ Y₂O₃/YSZ(110) in cross section. The Y₂O₃ film exhibits a well-ordered crystalline structure and a smooth surface, as expected from the streaky RHEED images in Fig. 4. The Fe overlayer, in contrast, is strongly islanded with a very rough, uneven surface. The large islands appear dark in the BF image, but a thin layer (approximately 35 nm thick) of Fe is visible between the islands. The weak contrast of this thin layer indicates that it is poorly ordered compared to the large Fe grains. The contrast in the dark field (DF) TEM image in Fig. 9(b) reveals that the Fe islands possess different crystalline orientations. The contrast variation in the center of the large Fe island is associated with slight orientation variations. Little diffraction contrast is observed in the Y₂O₃ film, consistent with the relatively narrow rocking curve collected for this sample (Fig. 2). The lower magnification STEM-HAADF image in Fig. 9(c) further corroborates that the material observed between the large Fe islands is also Fe, since it produces similar contrast in the image. In the high resolution STEM image shown in Fig. 9(d), the well-ordered single crystalline nature of the Y₂O₃ (110) film is clearly observed. A well-crystallized grain of Fe is present on the Y₂O₃(110) surface. From this single image, the orientation of the Fe grain cannot be determined, but it does not appear to possess an epitaxial relationship with the underlying Y₂O₃. Although the XRD pole figure presented in Fig. 4 and the EBSD data in Fig. 5 indicated that a fraction of the Fe film is epitaxially oriented, no epitaxial grains were found in the small region of the film presented in Fig. 9.

The effects of transmutation-produced He resulting from neutron irradiation in a fusion energy environment can be explored by implanting He⁺ in a material. The spatially-resolved He⁺ beam in a helium ion microscope can be utilized to explore He bubble formation in TEM-ready cross-sectional samples. Figure 10 presents cross-sectional TEM images of Fe/Y₂O₃/YSZ(110)

before (Fig. 10(a)) and after (Fig. 10(b)) 25 keV He⁺ irradiation with a fluence of $10^{22}/\text{m}^2$. In this region, no epitaxial Fe grains are present, so the Fe/Y₂O₃ interface is incoherent. In Fig. 10(b), He bubbles are present as dark spots in the bulk of the Fe and Y₂O₃ layers, as well as at the Fe/ Y₂O₃ interface. The He bubbles present at the Fe/ Y₂O₃ interface are significantly larger than those in the material bulk, as confirmed by the high resolution STEM image in Fig. 10(c). Analysis of electron energy loss spectra of similar bubbles corroborated the presence of He. In Fig. 10(d), very small bubbles with diameters of less than 1 nm are present in the Y₂O₃ lattice.

4. Summary

Thin films of Y₂O₃ were deposited on YSZ(100), (110), and (111) substrates by PLD. In all cases, epitaxial films were obtained, although the wide rocking curve widths indicated a significant defect structure in the films. Thin films of Fe were then deposited on the Y₂O₃/YSZ heterostructures by MBE. Structural analysis by RHEED, XRD, and RBS in the channeling geometry confirmed Y₂O₃ deposited on YSZ(111) was epitaxial but very rough, and the subsequent Fe film was purely polycrystalline. The Y₂O₃ film deposited on YSZ(100) was smooth and well-ordered, and the subsequent Fe film deposited with strong (110) texture. However, only very weak in-plane orientation was observed. In-plane disorder also reduced the RBS channeling yield from this sample. In contrast, the Fe film deposited on the smooth Y₂O₃/YSZ(110) possessed in-plane epitaxy and a reasonable RBS channeling minimum yield. The epitaxial orientation in these regions was determined to be Fe(211) **P** Y₂O₃(110) and Fe($\bar{1}11$) **P** Y₂O₃(001). Further analysis by S/TEM revealed that the Fe film consisted of large islands with non-epitaxial orientation; epitaxially-oriented grains are likely present but were not observed in the cross-sectional TEM image. Preliminary results of He⁺ implantation into a non-epitaxial region of Fe/ Y₂O₃/YSZ(110) revealed enhanced He bubble formation at the Fe/ Y₂O₃ interface relative to the respective matrices. Further optimization of the deposition parameters may increase the epitaxial fraction of Fe. Well-ordered crystallites of Fe with both epitaxial and non-epitaxial orientations on Y₂O₃ are a promising model system for high resolution S/TEM and/or

RBS channeling studies of the fundamental phenomena underlying the superior radiation tolerance of ODS steels and NFAs.

Acknowledgments

This work was supported by the U.S. Department of Energy (DOE), Office of Basic Energy Sciences, Division of Materials Sciences and Engineering. The experiments were carried out at the Environmental Molecular Sciences Laboratory (EMSL), a National Scientific User Facility located at Pacific Northwest National Laboratory (PNNL) and supported by the US DOE Office of Biological and Environmental Research. PNNL is a multiprogram national laboratory operated for DOE by Battelle.

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Figure captions

Fig. 1. High resolution XRD patterns for epitaxial Y_2O_3 thin films on (a) YSZ(100), (b) YSZ(110), and (c) YSZ(111). Insets in (b) and (c) show reciprocal space maps (RSMs) of the YSZ(331) and corresponding Y_2O_3 (662) reflections.

Fig. 2. High resolution XRD rocking curves of film and substrate for epitaxial Y_2O_3 films on YSZ(100), YSZ(110), and YSZ(111) substrates. FWHM values are indicated.

Fig. 3. (a) RHEED images of Y_2O_3 /YSZ(100) before and after Fe deposition. (b) Low resolution XRD of Fe/ Y_2O_3 /YSZ(100). (c) Fe(110) pole figure of Fe/ Y_2O_3 /YSZ(100).

Fig. 4. (a) RHEED images of Y_2O_3 /YSZ(110) before and after Fe deposition. (b) Low resolution XRD of Fe/ Y_2O_3 /YSZ(110). (c) Fe(110) pole figure of Fe/ Y_2O_3 /YSZ(110).

Fig. 5. EBSD inverse pole figure map (top) and pole figures generated from the map (bottom) of Fe on Y_2O_3 /YSZ(110).

Fig. 6. (a) RHEED images of Y_2O_3 /YSZ(111) before and after Fe deposition. (b) Low resolution XRD of Fe/ Y_2O_3 /YSZ(111). Asterisk indicates peak which could not be indexed. (c) Fe(110) pole figure of Fe/ Y_2O_3 /YSZ(111).

Fig. 7. RBS in the random and channeling directions for (a) Fe/ Y_2O_3 /YSZ(100), (b) Fe/ Y_2O_3 /YSZ(110), and (c) Fe/ Y_2O_3 /YSZ(111). Insets show the Fe backscattering peak in both geometries after background subtraction, and the resulting $\chi_{\min}$ value.

Fig. 8. (a) Schematic side view of Y_2O_3 (110) surface. Large blue spheres are O; small green spheres are Y. (b) Schematic side view of Fe(211) surface. (c) Schematic surface view of Fe(211)/ Y_2O_3 (110), indicating a possible epitaxial orientation. For clarity, only the Y surface net is shown for Y_2O_3 .

Fig. 9. Fe/ Y_2O_3 /YSZ(110) TEM ((a) and (b)) and STEM ((c) and (d)) micrographs.

Fig. 10. Fe/Y₂O₃/YSZ(110) TEM ((a) and (b)) and STEM ((c) and (d)) micrographs after irradiation with 25 keV He⁺ with a fluence of 10²²/m². He bubble formation is indicated by arrows in (c) and (d).

Fig. 1

Fig. 2

Fig. 3

Fig. 4

Fig. 5

Fig. 6

Fig. 7

Fig. 8

Fig. 9

Fig. 10