Author's Accepted Manuscript

www.elsevier.com

Structural and mechanical properties of γ-irradiated Zr/nb multilayer nanocomposites

M. Callisti, Sergio Lozano-Perez, T. Polcar

PII: S0167-577X(15)30702-3
DOI: http://dx.doi.org/10.1016/j.matlet.2015.10.057
Reference: MLBLUE19713

To appear in: *Materials Letters*

Received date: 16 September 2015
Accepted date: 12 October 2015

Cite this article as: M. Callisti, Sergio Lozano-Perez and T. Polcar, Structura and mechanical properties of γ-irradiated Zr/nb multilayer nanocomposites *Materials Letters,* http://dx.doi.org/10.1016/j.matlet.2015.10.057

Structural and mechanical properties of γ-irradiated Zr/Nb multilayer nanocomposites

M. Callisti [a,*], Sergio Lozano-Perez [b], T. Polcar [c]

[a] Materials Research Group, Faculty of Engineering and the Environment, University of Southampton, Southampton SO17 1BJ, UK

[b] Department of Materials, University of Oxford, Oxford OX1 3PH, UK

[c] National Centre for Advanced Tribology at Southampton, Faculty of Engineering and the Environment, University of Southampton, Southampton SO17 1BJ, UK

Corresponding author:
Materials Research Group, Faculty of Engineering and the Environment, University of Southampton, Southampton SO17 1BJ, UK
Building 7 / Room 4027 – Mailpoint M7
Southampton, Hampshire SO171BJ
United Kingdom
Tel.: +44 (0)23 8059 4438
* Corresponding author: M.Callisti@soton.ac.uk

Abstract

Zr/Nb multilayers with periodicities of 10, 30 and 60 nm were prepared by magnetron sputtering and irradiated for prolonged time (1311 h) by γ-rays with energy of 1.25 MeV and a dose of 510 kGy. A qualitative comparison between XRD patterns acquired before and after irradiation revealed a progressive increase of compressive stress, especially in Nb layers, for smaller periodicities with a consequent increase in hardness measured by nanoindentation. The combination of smaller grain size and radiation-induced defect density distribution, primarily in Nb layers, was found to be responsible for the observed radiation hardening effect.

Keywords: multilayers, irradiation embrittlement, scanning/transmission electron microscopy.

1. Introduction

Radiation-induced defects and corresponding changes in mechanical properties in ion- and neutron-irradiated materials have been thoroughly studied in the last decades on different material systems [1]. Whereas the effects of fast neutrons and helium bubbles formation are largely recognised, the impact of gamma (γ) ray damage to materials is generally not significant [2]. Nevertheless, when a large water gap exists as in reactor pressure vessels (RPVs), γ-rays damage is comparable or even higher to that produced by fast neutrons. Furthermore, due to the higher efficiency of γ-rays to produce freely-migrating defects, radiation-induced processes that depend on long-range defect fluxes to sinks (i.e. segregation to interfaces, precipitation) can be affected by γ-rays damage [2].

Interfaces in the form of grain boundaries and interphases are widely recognised for they healing effects against radiation-induced point defects, where recombination of vacancies and interstitials can take place [3-5]. Based on this concept, metals with diverse crystal structures (bbc, fcc, hcp) have been combined to fabricate multilayers with enhanced tolerance to radiation damage generated during He-ion and neutron bombardment [1]. Very little attention was paid regarding the effects of γ-rays on structure and mechanical properties of nuclear materials. It was reported that Nd-Fe-B alloys, widely used as magnets in radiation environments, amorphized after γ radiation at room temperature [6]. Hardening and structural disorder were also reported for Al alloys [7] and ferritic steels used in RPVs [8]. In nuclear power plants, Zr-based alloys are extensively used as cladding for nuclear core materials due to their ability to withstand reactor conditions for long periods of time [9], although radiation hardening and amorphisation are still of main concern. In this study we combine the excellent properties of Zr with the enhanced radiation tolerance of multilayer architectures in the form of Zr/Nb nanocomposites. Multilayers with different interface density distributions are produced and their structural properties and nano-mechanical behaviour investigated before and after γ-rays irradiation.

2. Experimental

Zr/Nb multilayers with different periodicities (λ) i.e. 10, 30 and 60 nm, were deposited by DC magnetron sputtering on single crystal (100) Si wafers with a total thickness ranging between 1.1 – 1.3 µm. Further details about the deposition process are reported elsewhere [10]. Gamma irradiation on the as-deposited multilayers was carried out in a ^{60}Co (1.25 MeV) γ-ray irradiation facility at ambient temperature. All samples were irradiated with the same dose of 510 kGy and a photon flux of ~ $2.4 \times 10^{10} cm^{-2} s^{-1}$ for a total irradiation time of 1311 h. Nanoindentation was employed to measure mechanical properties before and after irradiation by using a Berkovich indenter with loads between 3 – 4 mN. Loading and unloading time were fixed at 20 and 5 s, respectively, while a holding time at maximum load of 15 s was sufficient to saturate creep effects. Structural analyses were performed by grazing incidence X-ray diffraction (GIXRD) and Scanning transmission electron microscopy (STEM) with a JEOL ARM200F (cold-FEG) TEM at 200 kV. Lift-out cross-sectional TEM lamellae were prepared by focused ion beam (FIB).

3. Results and discussion

Fig. 1 shows the XRD patterns of the as-deposited Zr/Nb multilayers. Constituent elements exhibited a polycrystalline structure regardless of the periodicity. The inset in Fig. 1 shows the selected area electron diffraction (SAD) pattern acquired on the as-deposited multilayer with λ = 10 nm. Zr exhibited two main orientations, i.e. Zr (0002) and Zr ($10\bar{1}1$) parallel to the substrate, whereas Nb was oriented at random. As the periodicity was lowered down to 10 nm, a remarkable peak shift was observed especially for the Zr (0002) and satellite peaks appeared around the main diffraction planes. These features suggested the formation of a superlattice structure [11]. A similar crystallographic orientation was found for λ = 60 nm. Nb structure had a lateral grain size ranging between 2 – 5 nm and ~ 30 nm while for Zr it ranged between 4 – 7 nm and ~ 45 nm for λ = 10 and 60 nm, respectively. Further details about structural and mechanical properties of as-deposited Zr/Nb multilayers are reported elsewhere [10].

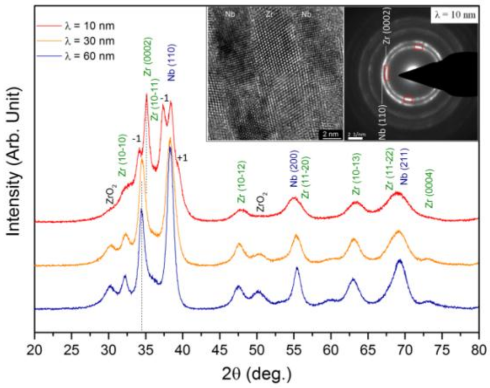

Fig. 1

In order to shed light about the effects of γ-ray irradiation on potential microstructural changes, a qualitative comparison between XRD patterns acquired before and after irradiation was carried out (Fig. 2). In the as-deposited multilayers as compared to the stress-free reference standards, Nb was found to be under compressive stress decreasing for larger periodicities, whereas Zr was under compressive stress except for λ = 10 nm, where tensile stress generated likely due to the formation of a superlattice structure.

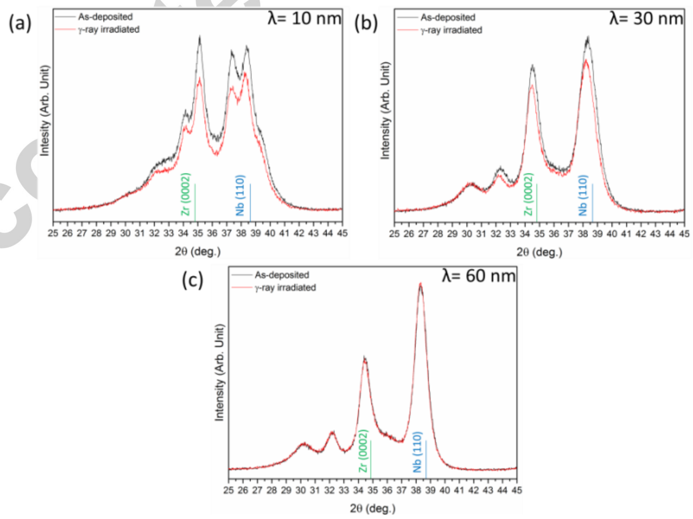

Fig. 2

After irradiation, no phase changes for the constituent elements were found; however, changes in peaks position were observed. In particular, irradiation had only limited effects on Zr structure, while Nb pattern gradually shifted toward lower 2θ angles for smaller periodicities.

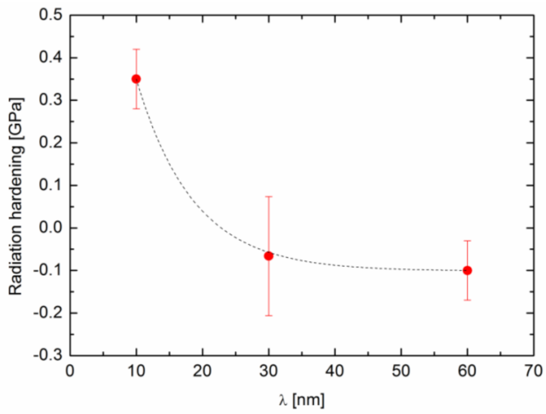

Fig. 3

Fig. 3 shows the radiation hardening (ΔH = $H_{\gamma-ray} - H_{As-dep.}$) in relation to periodicity. A neat hardening effect was found for the smallest periodicity. In the case under investigation, Compton scattering is the most probable phenomenon to occur between γ-rays and target metals. This mechanism consists of a direct interaction between γ-ray and outer, less tightly bounded electrons, resulting in the production of fast electrons. The latter interact with lattice atoms generating primary knock-on atoms (PKAs) with consequent formation of displacement cascades and structural disorder in the form of vacancies, interstitials and dislocations [12]. For the Compton scattering, the linear attenuation coefficient, which accounts for the fraction of γ-rays being absorbed by the target material, strongly depends on the density (ρ) of the absorber [12]. In the case under study, $\rho_{Nb} \approx 1.3 \cdot \rho_{Zr}$, which suggests that a larger damage has to be expected in Nb layers. Fig. 2 shows that variation in compressive residual stress is always larger in Nb rather than in Zr structure, with a certain dependency on the periodicity. These results suggest that a more significant structural disorder is generated in Nb layers under γ radiation.

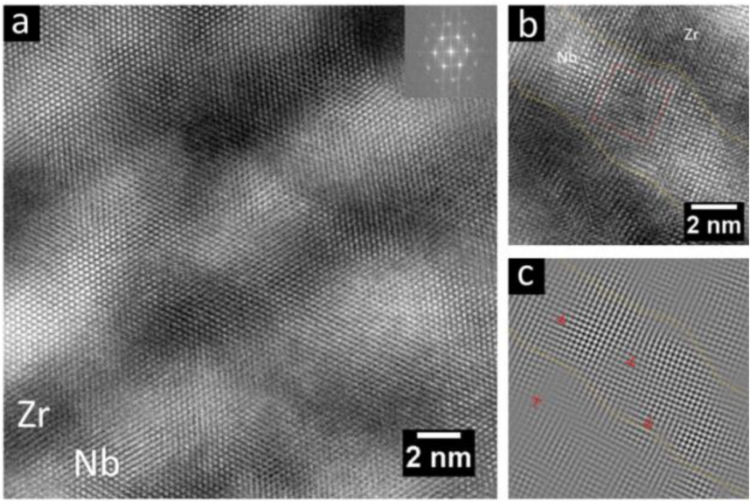

Fig. 4

Fig. 4a shows that the layered structure for λ = 10 nm was retained after irradiation; there was no evidence of post-irradiation interfacial mixing across the multilayer. Moreover, irradiation did not change the crystal structure and orientations of grains within Zr and Nb layers. Local disorder in the form of dislocations, formed away from interfaces, and consequent atomic planes bending was observed mostly within Nb layers (Fig. 4 b – c) of the irradiated sample with respect to its as-deposited condition (inset in Fig. 1). Notwithstanding the higher interface density distribution for λ = 10 nm, radiation-induced defects and consequent structural disorder persisted as suggested by the increase in compressive residual stress. A similar scenario was expected for the other multilayers with larger periodicities (larger grain size), where annihilation of point defects is even more difficult owing to the longer distance that point defects have to cover to reach grain boundaries and interphases. The hardening effect (Fig. 3) observed for the multilayer with λ = 10 nm is attributed to the combined effect of radiation-induced defects, especially in Nb layers, and smaller grain size. In particular, dislocation gliding is more difficult when defects are formed in smaller grains, where dislocations barely can deflect their path around point defects or move across strain fields, due to the constraining effects of surrounding interphases and grain boundaries. An average strain field of ~ 9 nm^2 was measured around single dislocations in Nb layers (λ=10 nm), which covers distances close to the average grain size. On the other hand, for larger periodicities (larger grain size) the presence of defects has a minor impact on limiting dislocation gliding; therefore, hardening effect is absent or negligible. As a result, γ-ray irradiated multilayers with larger periodicities can still accommodate similar amounts of deformation as in as-deposited state.

4. Conclusions

The effects of γ-ray irradiation on the structural and mechanical properties of Zr/Nb multilayers with different periodicities were investigated by combination of nanomechanical testing and structural analyses. Nb layers underwent a larger increase in compressive residual stress for lower periodicities due to formation of radiation-induced structural disorder. The combination of smaller grain size and radiation defects was found to be responsible for the observed hardening effect. In spite of the radiation-induced defects, large grains still allow dislocation gliding as easily as in pristine (i.e. non-irradiated) grains; therefore, multilayers with larger periodicities are found to exhibit a superior structural integrity and mechanical performance in γ-radiation environments.

Acknowledgments

The South of England Analytical Electron Microscope (EPSRC grant code EP/K040375/1) is acknowledged for access to the electron microscopy facilities.

References

[1] I.J. Beyerlein, M.J. Demkowicz, A. Misra, B.P. Uberuaga, Progress in Materials Science 74 (2015) 125-210.

[2] J. Kwon, A. T. Motta, Annals of Nuclear Energy 27 (2000) 1627-1642.

[3] W. Z. Han, M. J. Demkowicz, E. G. Fu, Y. Q. Wang, A. Misra, Acta Materialia 60 (2012) 6341-6351.

[4] P.A. Thorsen, J. B. Bilde-Sorensen, B. N. Singh, Scripta Materialia 51 (2004) 557-560.

[5] K. Hatter, M. J. Demkowicz, A. Misra, I. M. Robertson, R. G. Hoagland, Scripta Materialia 58 (2008) 541-544.

[6] Y. C. Han, S. Lee, H. W. Kwon, Y. S. Kang, Nuclear Instruments and Methods in Physics Research B 260 (2007) 599-602.

[7] G. Saad, S. A. Fayek, A. Fawzy, H. N. Soliman, E. Nassr, Materials Science & Engineering A 607 (2014) 132-137.

[8] J. F. Knott, C. A. English, D. R. Weaver, D. P. G. Lidbury, International Journal of Pressure Vessels and Piping 82 (2005) 929-940.

[9] S. I. Choi, J. H. Kim, Nuclear Engineering and Technology Vol. 45 N. 3 June 2013.

[10] E. Frutos, M. Callisti, M. Karlik, T. Polcar, Materials Science & Engineering A 632 (2015) 137-146.

[11] Y. Lui, D. Bufford, H. Wang, C. Sun, X. Zhang, Acta Materialia 59 (2011) 1924-1933.

[12] Practical Gamma-ray Spectrometry, 2nd Edition, Gordon R. Gilmore, ISBN 978-0-470-86196-7, Published by Wiley-VCH Verlag, Wenheim, Germany, 2008 01/2008.

Figure captions

Fig. 1 – XRD patterns of the as-deposited Zr/Nb multilayers. The insets shows a HR-TEM image and the SAD pattern for the multilayer with λ=10 nm.

Fig. 2 – XRD analyses of the multilayers before and after irradiation in relation to the periodicity: (a) 10 nm, (b) 30 nm, (c) 60 nm.

Fig. 3 – Radiation hardening in relation to the periodicity. Dashed line is reported as guideline.

Fig. 4 – HR-HAADF STEM images for λ = 10 nm after γ irradiation: (a) layered structure with Zr($10\bar{1}1$)//Nb(110), (b) strain field around edge dislocations in the layers, (c) Fourier filtered image of (b) with highlighted edge dislocations and interfaces.

Research highlights:

- PVD Zr/Nb multilayers with different periodicities were synthesised.
- Possible effects of γ-radiation on the structure of Zr/Nb multilayers are proposed.

- A more pronounced radiation hardening effect was found on finer grain structures.