5.2 High Neutron Dose Irradiation of Dielectric Mirrors — K. J. Leonard, G. E. Jellison Jr., N. A. P. K. Kumar and L. L. Snead (Oak Ridge National Laboratory)

OBJECTIVE

The goal of this work is to evaluate the upper neutron irradiation dose and thermal limits of two promising dielectric mirror types, through an investigation of the radiation and thermally induced microstructural and optical property changes. While specifically of interest for use in laser control of inertial confinement fusion systems, the examination of the radiation induced structural changes in the films will be beneficial for the development of other thin-film based electronic components and sensors used in nuclear applications.

SUMMARY

During this semiannual reporting period, work was completed on low dose (0.001 to 0.1 dpa) examinations culminating in the presentation of data at the Radiation Effects in Insulators conference in Helsinki, Finland in July 2013. Publication of the paper "The role of microstructure on the optical performance of neutron irradiated dielectric mirrors" has been confirmed for the Journal of Nuclear Materials in 2014, volume 445, pages 281-290. The optical property characterization of the 1 and 4 dpa-irradiated mirrors has also been completed, with data presented in poster form at the International Conference of Fusion Reactor Materials in Beijing, China in October. This semiannual contribution briefly summarizes the low dose results, which are presented in more detail in the previous contributions, as well as the optical property data for the high dose samples.

PROGRESS AND STATUS

Failure Mechanisms in Dielectric Mirrors

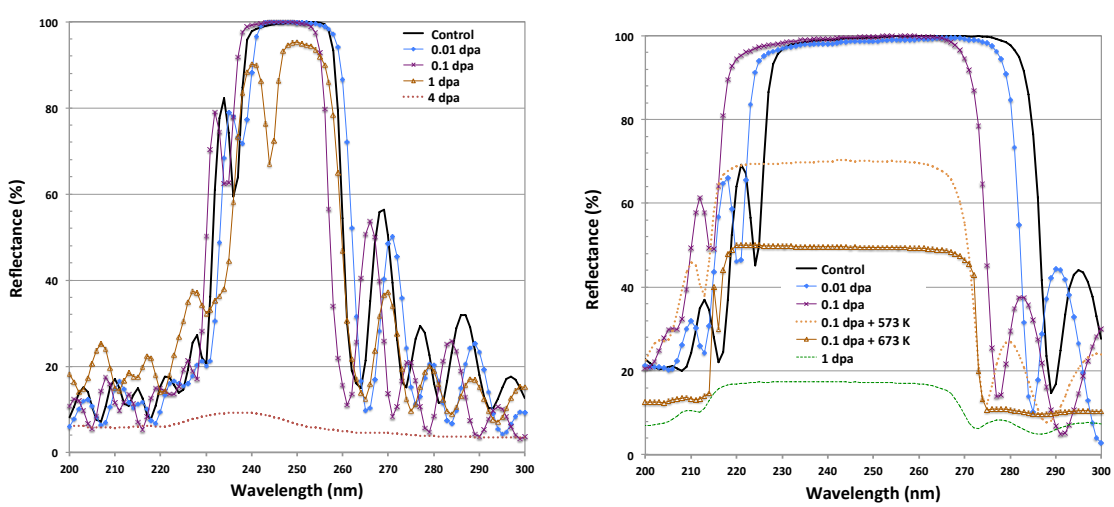
The beginning of this semiannual period saw the completion of the low dose (0.001 to 0.1 dpa) characterization work on the Al_2O_3/SiO_2 and HfO_2/SiO_2 dielectric mirrors. Failure through film delamination resulting in loss in reflectance only occurred following post-irradiation annealing of the 0.1 dpa HfO_2/SiO_2 mirror. However, considerable interdiffusion of Al and Si was measured across the film interfaces of the Al_2O_3/SiO_2 mirror that may be related to reflectance losses at higher (1 dpa and greater) displacement damage levels.

Reflectance loss in the HfO_2/SiO_2 dielectric mirror observed following 1 hr. annealing treatments at 573 and 673 K of the 0.1 dpa irradiated samples was caused by delamination of the films by defects generated at the HfO_2 film/sapphire substrate interface. These defects are formed from the buckling of the first HfO_2 film layer (the mirror consists of alternating HfO_2 and SiO_2 layers with an HfO_2 being the first and last film layers in the 11 bi-layer stack) as a result of the loss of adhesion to the sapphire substrate due to the amorphization of the sapphire in the vicinity of the film interface. The defects at the interface produce a higher state of stress in the films during cooling from the annealing temperatures, which upon reaching a critical stress value can create localized delamination of the entire film stack. The loss in reflectance is greater with

increasing annealing temperature and is suspected to be the source of failure appearing temperature.

Optical Properties of High Dose Neutron Irradiated Mirrors

A comparison of the spectral reflectance curves for the two mirror types following select irradiation and post-irradiation exposures is shown in Figure 1. Both mirrors were designed for peak reflectivity at 248 nm. For the Al_2O_3/SiO_2 mirrors irradiation to 1 dpa resulted in a decrease in the working range (the width of peak reflectance shown in the curves in Figure 1) of the mirror, with a reduction in reflectance to 95% at 248 nm. Furthermore, the working range is also shifted to longer wavelengths. The change in shape of the reflectance spectra is likely due to the formation of secondary phases between the constituent film types. However, the loss in reflectance is through delamination, likely from the secondary phase particles that produce stress concentrators in the film stack. Microstructural analysis of these films is underway. Complete loss in reflectance is observed in the Al_2O_3/SiO_2 mirrors following 4-dpa irradiation. Signs of film delamination could be observed visually following irradiation. Figure 2 compares the visual changes in the Al_2O_3/SiO_2 mirror with increasing irradiation leading to the darkening in the optical range as well as the appearance of delaminated regions (speckling) occurring at higher levels of dpa. Similar changes in appearance are observed for the HfO_2/SiO_2 mirror. The small difference in surface delamination between the 1 and 4 dpa samples suggests that failure is very rapid in this range.



(a) (b)

Figure 1. Photo spectrometry results of the (a) Al_2O_3/SiO_2 and (b) HfO_2/SiO_2 mirrors in the unirradiated and select irradiation and post-irradiation annealed conditions. The reflectivity data is normalized based on the absolute reflectivity measurements conducted through direct laser reflectance measurements.

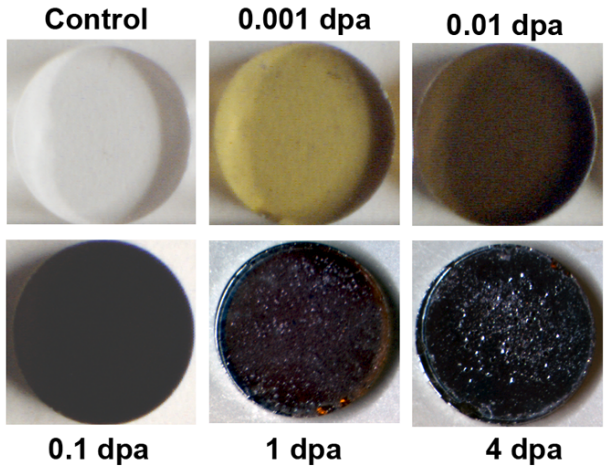


Figure 2. Visual changes in the Al_2O_3/SiO_2 mirror with increasing neutron irradiation. Similar changes are observed for the HfO_2/SiO_2 mirror.

The HfO_2/SiO_2 mirror showed reflectance values below 20% following 1 dpa exposure (Figure 1b) and no measureable reflectance following 4 dpa. The mechanisms responsible for the losses are presumably an extension of those identified in microstructural characterization of the lower dose samples. As discussed in the previous section, partial amorphization of the sapphire near the HfO_2 film interface occurs with irradiation produced a 20 nm thick layer following 0.1 dpa. It is expected that with higher displacement damage the amorphous region has further weakened the interface leading to the buckling of the HfO_2 without the need for a post-irradiation anneal. Cooling from the irradiation temperature would therefore be enough to cause elevated stresses at these defects to cause delamination.

Discussion

The work conducted in the past year on the Al_2O_3/SiO_2 and HfO_2/SiO_2 dielectric mirrors provided detailed insight into both the upper neutron irradiation dose and thermal exposure limits of mirrors. Thermal exposures up to 675 K produced similar changes in the materials as compared to neutron irradiation to 0.1 dpa at 448 K. It is likely that these mirrors could be capable of higher temperatures for short durations, but they are going to be very sensitive to the rate of cooling as well as the amount of accumulated damage present in the material either through thermal or displacive radiation exposure.

The Al_2O_3/SiO_2 mirror shows greater stability of the two mirror types studied despite the significant interdiffusion of Al and Si between the constituent layers. The 10% loss in reflectance following 1 dpa exposure is small in comparison to losses observed in the HfO_2/SiO_2 mirrors under equivalent conditions. Furthermore, our earlier work demonstrated better laser damage threshold values for the Al_2O_3/SiO_2 mirror [1]. This is likely influenced by the closer matching of properties of the amorphous Al_2O_3 and SiO_2 layers of the mirror, as well as the near atomic level smoothness of the deposited layers

as compared to that of the HfO_2 layers in the other mirror. Despite some decreases in roughness with irradiation of the HfO_2 layers, the large microstructural changes associated with film densification, phase changes in HfO_2 and amorphization near the substrate/film interface would only lead to further deterioration of laser damage tolerance.

From a design perspective, the Al_2O_3/SiO_2 mirror offers a stable platform for systems in fusion applications where laser based diagnostic devices are used. The peak reflectance for the mirror can be tuned to different wavelengths by varying the thickness of the constituent films. There may be some limitation if substantially thick layers are required and ion-beam assisted vapor deposition can no longer maintain an amorphous, smooth and defect free films. Due to the large difference in refractive indices of the constituent layers the HfO_2/SiO_2 mirror offers increased working range of the mirror with fewer number of bi-layers required (better control of defects) for high reflectance. However, the lack of radiation stability of the HfO_2 layer makes this mirror less attractive for nuclear applications. Other materials with large refractive index values compared to SiO_2, such as TiO_2 and ZrO_2 also suffer from similar radiation-induced phase changes as HfO_2.

The evaluation of other film layer combinations may lead to some improvements over the Al_2O_3/SiO_2 mirror, though these improvements are likely to be small. However, the Al_2O_3/SiO_2 mirror maintained ≥ 95% reflectance up to 1 dpa neutron exposure, which is two orders of magnitude greater than for the laser control systems of the inertial confinement fusion system that this study was initially intended to examine. The outcomes of this work definitively showed the link between microstructural changes in the mirrors to that of its optical properties and identified the mechanisms leading to delamination of film layers.

Current and Future Work

Work on the 1 and 4 dpa mirrors is completing with the examination of the microstructures through transmission electron microscopy. The key issues that will be examined in these samples are 1.) If secondary phase development occurred at the Al_2O_3/SiO_2 film interfaces and, 2.) If the delamination mechanisms in the HfO_2/SiO_2 mirror following 1 and 4 dpa are the same as that observed in the 0.1-dpa mirrors following post-irradiation annealing.

Further work on optical properties of diagnostic system based materials will be applied to the identification of needed areas of examination or clarifications related to materials behavior in fusion environments. The purpose of which is to identify possible areas in which the US Fusion Materials Program could be situated in order to provide greater international leadership in the area of diagnostic systems.

REFERENCES

[1] L. L. Snead, K. J. Leonard, G. E. Jellison Jr., M. Sawan and T. Lehecka, *Fusion Sci. Technol.* **56** (2), 1069 (2009).