

Available online at www.sciencedirect.com



Journal of Nuclear Materials 347 (2005) 255-265

journal of nuclear materials

www.elsevier.com/locate/jnucmat

Effect of multi-shot X-ray exposures in IFE armor materials

J.F. Latkowski^{a,*}, R.P. Abbott^{a,b}, R.C. Schmitt^{a,b}, B.K. Bell^a

^a Lawrence Livermore National Laboratory, 7000 East Avenue, Mailstop L-641, Livermore, CA 94551, USA ^b University of California at Berkeley, 4155 Etcheverry Hall, MC 1730, Berkeley, CA 94720, USA

Abstract

As part of the High Average Power Laser (HAPL) program the performance of tungsten as an armor material is being studied. While the armor would be exposed to neutrons, X-rays and ions within an inertial fusion energy (IFE) power plant, the thermomechanical effects are believed to dominate. Using a pulsed X-ray source, long-term exposures of tungsten have been completed at fluences that are of interest for the IFE application. Modeling is used in conjunction with experiments on the XAPPER X-ray damage facility in an effort to recreate the effects that would be expected in an operating IFE power plant. X-ray exposures have been completed for a variety of X-ray fluences and number of shots. Analysis of the samples suggests that surface roughening has a threshold that is very close to the fluences that reproduce the peak temperatures expected in an IFE armor material.

© 2005 Elsevier B.V. All rights reserved.

1. Introduction

While it is relatively easy to design components to survive against single-shot damage such as melting, cracking and ablation, this is inadequate when designing an inertial fusion energy (IFE) target chamber. IFE power plants are expected to operate at repetition rates of 5–10 Hz. For a system availability of 85%, this implies 134–268 million shots per year. At 5 Hz, even 0.1 nm of material loss per-shot would be more than 1 cm of material removed per year. Clearly, this is not acceptable.

Thermomechanical effects can lead to changes in the surface characteristics of a material such as roughening. Roughening is a concern in that it can lead to large gradients in the stress, which can cause cracking and/or enhance the growth rates of preexisting cracks.

Further, thresholds for single-shot damage are always far greater than those for many shots. For example, Zaghloul, Tillack and Mau [1] have shown that the laser-induced damage fluence for aluminum drops from $\sim 140 \text{ J/cm}^2$ for a single shot to $<20 \text{ J/cm}^2$ for 10^5 shots. Projections give a damage limit of 5–8 J/cm² for 10^8 shots. It seems reasonable to assume that a similar decrease in the damage threshold may be observed for X-rays as well. Therefore, it is crucial to conduct experiments at a high repetition rate in order to provide a sufficient number of shots and capture such effects. An experimental repetition rate of 10 Hz means that exposures of 10^5 shots can

^{*} Corresponding author. Tel.: +1 925 423 9378; fax: +1 925 423 4606.

E-mail address: latkowski@llnl.gov (J.F. Latkowski).

^{0022-3115/\$ -} see front matter @ 2005 Elsevier B.V. All rights reserved. doi:10.1016/j.jnucmat.2005.08.018

be completed on a routine basis, while 10^6 shots is a reasonable upper limit.

Much of the radiation released from an IFE target will be in the form of high-energy neutrons, which will not significantly heat the armor material. The remaining energy is in the form of charged particles and X-rays. Only $\sim 1.4\%$ of the target output is released as X-rays, but additional X-rays may be produced via reverse Bremsstrahlung as lowenergy ions stop in the chamber gas, if any. The vast majority of the armor heating is caused by charged particles. Output spectra from various IFE target designs have been calculated by Perkins [2] and can be found at the link given in Ref. [3]. Details of the so-called 'threat spectra' and its interaction with the armor in an IFE power plant are given by Raffray et al., in Ref. [4]. In addition, considerable work on this matter has been completed by the team at the University of Wisconsin [5]. Although a wide variety of particle types and energies would be incident upon an IFE first wall, many of the effects are thought to be thermomechanical in nature [6]. As such, an X-ray source is capable of serving as an exposure simulator. In the present work, the XAPPER X-ray damage facility is used for this purpose. XAPPER is based upon an extreme ultraviolet (EUV) X-ray source developed and produced by PLEX LLC [7].

2. Experimental capabilities

The XAPPER X-ray damage experiment utilizes an EUV source designed and built by PLEX LLC of Cambridge, Massachusetts [7]. The source operates via a plasma pinch. While the source is capable of operation with argon and nitrogen plasmas, the source output with these gases is significantly lower, and thus, only xenon plasmas have been used to date on XAPPER. Fig. 1 shows a typical X-ray spectrum captured using a McPherson grazing incidence spectrometer. The X-ray spectrum can be modified by varying the gas pressure within the pinch, the discharge voltage, or by filtering the output prior to striking the sample.

The source output in the EUV is approximately 0.25 J/sr with a full-width at half-maximum (FWHM) pulse length of ~40 ns . For a sample sitting at 30 cm from the plasma head, this translates into a per-shot X-ray fluence of $<0.3 \text{ mJ/cm}^2$. In order to provide a much higher X-ray fluence as well as mitigate debris contamination from the plasma pinch, XAPPER utilizes an ellipsoidal focusing optic. The system layout used in tungsten exposures is indicated in Fig. 2. Output from the source is restricted to the range of angles that intercept the focusing optic. This is accomplished using a foil comb, which is shown in Fig. 3. The foil comb



Fig. 1. The XAPPER X-ray spectrum ranges from ~80 to 140 eV.



Fig. 2. XAPPER experiments utilize an ellipsoidal focusing optic to obtain increased X-ray fluences.



Fig. 3. The foil comb reduces debris and restricts X-ray output to those angles that intercept the focusing optic.

not only restricts X-ray output to that which intercepts the optic, but it also significantly reduces the amount of debris leaving the plasma head.

To measure the X-ray per-shot fluence at the sample plane, a series of measurements is completed prior to each exposure. First, the X-ray power is measured using a vacuum calorimeter from Scientech, Inc. Second, the focused X-ray beam is imaged onto a Princeton Instruments charged coupled



Fig. 4. The focused X-ray beam has a spot size of ~ 1 mm.

device (CCD) camera through a 4-µm-thick zirconium foil. The zirconium filter reduces the EUV fluence by $\sim 10^5$, thereby protecting the CCD camera from severe damage. Fig. 4 is an image of a typical X-ray spot with a FWHM spot size of ~ 1 mm. By integrating the background-corrected CCD image, one is able to determine the effective number of counts per unit energy. Given the counts in the peak channel and the size of the pixel, one obtains the average peak X-ray fluence per-shot. Since the calorimeter is used to determine the power in an unfocused beam, diagnostics are not damaged. Since the CCD measurement is only used to provide an X-ray profile, the measurements are insensitive to the uncertainties in the zirconium filter thickness.

Shot-to-shot variations in the spot location and X-ray fluence are manageable. CCD measurements indicate that the location of the peak intensity has a standard deviation of better than 70 μ m in an overall spot size of ~1 mm. The peak X-ray fluence has a standard deviation of ~11% of the target fluence. Variations in the X-ray fluence are due primarily to source output variability, as indicated by direct photodiode energy measurements.

XAPPER can be operated at repetition rates of up to 10 Hz. Thus, an exposure of 10^5 pulses requires less than 3 h to complete. To date, the longest exposures completed have been 2×10^5 pulses on aluminum mirrors. Tungsten samples have been exposed for as many as 10^5 pulses. XAPPER can be operated for millions of pulses prior to routine maintenance of the plasma head.

3. Modeling

Computational support is necessary to direct the experimental activities. Modeling of the time-temperature history experienced by an IFE armor is accomplished using the RadHeat finite difference code. RadHeat is able to determine the transient temperature evolution of multi-layer (and material) targets that are exposed to any number of photon and ion spectra. RadHeat allows arbitrary convective cooling (or heating) at the front and back surface, as well as allowing radiation transport with fixed temperature surroundings. RadHeat utilizes an implicit numerical scheme, which leads to increased stability and a faster solution. Additional details regarding RadHeat may be found in Ref. [8].

Using the Perkins target output spectrum for the 154 MJ yield target, heating of the tungsten armor has been calculated. Numerical effects resulting from the ion Bragg peak require one to interpolate between the energy groups provided by Perkins. Typically, each energy bin must be subdivided into as many as 20 separate bins. Both Raffray and Peterson have reported similar effects.

The prompt X-ray pulse is assumed to be a 1 ns square pulse, while the various ions are all assumed to have a 0.1 ns pulse length. Vacuum conditions are assumed for the 6.5-m radius chamber, and time-of-flight spreading of the ion pulses occurs during transit across the vacuum. A back surface heat transfer coefficient of 10 kW/m2-K is assumed. Fig. 5 shows the surface temperature as a function of time. Note that, due to the logarithmic scale, the intra-pulse details only can be seen for the first of 10 pulses. At ~27 ns, a modest spike in the tem-



Fig. 5. RadHeat results for heating of the tungsten armor show a peak surface temperature of 3256 K for the first pulse and 3307 K for the tenth pulse.

perature is seen resulting from the prompt X-ray heating. This is followed by the arrival of the burn ions at $\sim 1 \mu$ s, and the debris ions at $\sim 1.5 \mu$ s. For the first pulse, a peak surface temperature of 3256 K is calculated. This is similar to results reported by Raffray [4]. Over the first 10 pulses (at 10 Hz), the peak temperature ratchets up to 3307 K. The armor has not yet achieved thermal equilibrium in an average power sense. This should occur in the first couple hundred pulses. As shown in Fig. 6, calculations using the XAPPER X-ray spectrum show that a fluence of $\sim 0.8 \text{ J/cm}^2$ is able to replicate the same peak surface temperature for a sample starting at an initial temperature of 773 K.

Although 0.8 J/cm^2 matches the expected peak surface temperature for the IFE case described above, other fluences are also of interest. Table 1 lists X-ray fluences used on XAPPER for the exposures presented in this paper. For each fluence, the corresponding peak surface temperature (for the first pulse) is indicated as well. In Table 1 an initial temperature of 300 K is assumed (as opposed to 773 K, which is discussed above). XAPPER X-ray exposures have been completed with fluences of



Fig. 6. XAPPER can match the expected armor surface temperature with an X-ray fluence of $\sim 0.8 \text{ J/cm}^2$.

Table 1

X-ray fluences used on XAPPER and the corresponding peak surface temperatures for tungsten armor at an initial temperature of 300 K

X-ray fluence (J/cm ²)	Peak surface temperature (K)
0.5	1840
0.7	2470
0.8	2780 (3250 K for $T_0 = 773$ K)
1.0	3385
1.2	Melts (>3695 K) @ 33 ns
	into 40 ns pulse

0.5, 0.7, \sim 1.0 and \sim 1.2 J/cm². The \sim 1.0 and \sim 1.2 J/ cm² values are approximate, as those particular data



Fig. 7. The tungsten thermal conductivity varies as a function of temperature.



Fig. 8. The tungsten heat capacity varies with temperature. Note that values above 3273 K have been set to 200 J/kg-K.

sets were taken prior to development of the calorimeter/CCD method for measuring the X-ray fluence. The fluence values are based upon anecdotal evidence, and thus, are suspect to considerably larger error bars.

Temperature-dependent properties for tungsten have been taken from the ARIES web site [9]. Note that a couple of minor changes are made to the basic properties. Specifically, the thermal conductivity has been set to 70 W/m-K for temperatures greater than or equal to 3773 K. Also, the heat capacity has been set to 200 J/kg-K for temperatures greater than or equal to 3273 K [10]. These modifications are reasonable given the significant scatter in the available data at such high temperatures. They have a relatively minor effect upon the modeling results. X-ray opacities for pure tungsten are taken from the Center for X-ray Optics web site [11]. Figs. 7–9 show plots of the data used for these important parameters.

4. Tungsten exposures on XAPPER

Both powder metallurgical (powder met) and single crystal tungsten samples have been exposed to a variety of X-ray fluences and for various numbers of pulses. Samples were provided by Dr. Lance Snead of Oak Ridge National Laboratory. The X-ray fluences have ranged from 0.5 to $\sim 1.2 \text{ J/cm}^2$, and the number of pulses has ranged from singleshot exposures to as many as 10^5 pulses in a given



Fig. 9. The X-ray opacity varies as a function of the photon energy.

location. A different sample of each type was used at each separate fluence. All samples were irradiated beginning at room temperature, but the average X-ray power causes the minimum temperature to 'ratchet up' by 200–300°. This is shown in Fig. 10.

Tungsten samples have been analyzed using a Veeco white-light interferometer. Pre-irradiation measurements typically show initial surface roughness of ~ 20 nm for powder met and ~ 10 nm for the single crystal samples. Visual inspection of samples is performed as well. Imaging has been done with a simple digital camera as well as through a low-power microscope. Fig. 11 shows images of a single crystal sample that was shot at 0.5 J/cm^2 alongside a powder met sample that was shot with 0.7 J/cm^2 . In both cases, a slight discoloration is visible in the location that received 10^5 pulses. Discoloration was also observed in the powder met

sample shot at 0.5 J/cm^2 and the single crystal sample shot at 0.7 J/cm^2 . In none of these four lower-fluence samples is there obvious damage at the sites that received fewer than 10^5 pulses.

Fig. 12 shows a powder met sample that was severely damaged from the $\sim 1.2 \text{ J/cm}^2 \text{ X-ray}$ fluence. Single-shot damage is evident in this sample. This agrees with our model, which predicts that single-shot melting will occur at $>1 \text{ J/cm}^2$.

Prior to irradiation the single crystal tungsten was quite smooth. Twenty-four measurements from areas throughout the sample, each $240 \times 180 \ \mu\text{m}$ in size, give a surface roughness of $7.7 \pm 1.7 \ \text{nm}$. The powder met sample was a bit rougher, with a surface roughness of $16 \pm 1.8 \ \text{nm}$. When irradiated for as many as 10^5 pulses at $0.5 \ \text{J/cm}^2$, neither sample showed any signs of statistically significant roughening. Fig. 13(a) and (b) show a white-light



Fig. 10. When XAPPER is operated at 10 Hz, the average X-ray power causes the minimum temperature to increase from 300 K to as much as 550 K.



Fig. 11. In the samples shot at X-ray fluences of 0.5 and 0.7 J/cm², subtle damage is visible at the location that received 10⁵ pulses.



Fig. 12. Severe damage is observed in this powder met tungsten sample that was shot with an X-ray fluence of $\sim 1.2 \text{ J/cm}^2$.

interferometer (WLI) scan of the powder met sample. The scan was taken in the region where visible discoloration of the sample occurred. The powder met surface roughness over about 1 mm^2 area increased from 17 to 19 nm after 10^5 pulses. This is not statistically significant. Similar results are observed for the single crystal sample.

When the X-ray fluence is increased to 0.7 J/cm², little or no change occurs in either the single crystal or powder met samples. As Fig. 14 shows, the single crystal sample actually may have been smoothed by a small amount. However, the changes do not appear to be statistically significant.

At an X-ray fluence of $\sim 1.0 \text{ J/cm}^2$, some surface roughening is observed. Prior to irradiation, the single crystal roughness was 10 nm, while the powder met was 20 nm. Fig. 15 shows pre- and postirradiation WLI images for the powder met tungsten. After 10⁴ pulses, the roughness increased to 72 nm. Interestingly, the single crystal reaches the same roughness after 30000 pulses at the same fluence. Fig. 16 summarizes the $\sim 1.0 \text{ J/cm}^2$ irradiations. Note that the single crystal roughening appears to be retarded in that it does not begin until sometime between 10⁴ and 3×10^4 pulses. This suggests that there may be a threshold below which roughening occurs. It is unclear what effect might



Fig. 13. The surface roughness of powder met tungsten exposed to 10^5 pulses at 0.5 J/cm² is essentially unchanged.



Fig. 14. Single crystal tungsten may experience some smoothing when exposed to 10⁵ pulses at 0.7 J/cm².



Fig. 15. Powder met tungsten undergoes roughening when exposed to 10^4 pulses at ~ 1.0 J/cm². Note that the height scales differ in the pre- and post-irradiation WLI images.

cause such behavior. It would be interesting to continue the powder met tungsten exposures beyond 10^4 pulses, as well as taking single crystal tungsten beyond 3×10^4 pulses. It would be interesting to learn whether or not the roughening is linear between numbers of pulses (e.g., between 10^4 and



Fig. 16. Surface roughness in powder met and single crystal tungsten grows with an increasing number of pulses at ~ 1.0 J/cm².

 3×10^4 pulses). Also, it would be interesting to learn whether or not the roughness continues to increase out to a larger number of pulses.

For the samples irradiated at a fluence of $\sim 1.2 \text{ J/cm}^2$, a very different result was observed. As Fig. 12 shows, severe damage was observed in powder met tungsten, with similar results in the single crystal sample. Given how bad the powder met sample looks to the eye, one might expect that the roughening was catastrophic. This is, in fact, the case. As shown in Table 1, tungsten should melt at ~ 33 ns into a 40-ns-long 1.2 J/cm² X-ray pulse. This is consistent with the single-shot damage observed in Fig. 12.

Given the severity of the visible damage, it is not surprising to learn that the surface has been roughened significantly. Prior to irradiation, this powder met tungsten sample had a surface roughness of 33 nm. With only a single shot at $\sim 1.2 \text{ J/cm}^2$, the roughness increased to 290 nm. Fig. 17 shows the WLI measurement results for the spot that received 3000 pulses. Note that the sample is extremely cratered and that portions of the data are missing due to the interferometer's inability to measure at such steep angles within the crater. Clearly, this sample indicates that tungsten could not survive for any reasonable length of time at such high Xray fluences. This would appear to refute hopes by



Fig. 17. The powder met sample exposed to 3000 pulses at $\sim 1.2 \text{ J/cm}^2$ is damaged so severely that the interferometer is unable to collect data within the crater.

some that it might be acceptable to allow the surface to melt and re-solidify each pulse.

While the $\sim 1.2 \text{ J/cm}^2$ results are disappointing, they certainly are not surprising. Calculations predict single-shot melting at this fluence, and it far exceeds the expected operating point within an IFE power plant. This is not a disturbing result and in no way causes alarm regarding the design of the tungsten armor for IFE.

5. Conclusions and future work

The series of experiments reported herein provide bounding circumstances regarding the roughening of various tungsten samples. At X-ray fluences of 0.5 and 0.7 J/cm², no statistically significant surface roughening is observed for either powder met or single crystal tungsten. In fact, it is possible that the single crystal tungsten was somewhat smoothed at a fluence of 0.7 J/cm². At a fluence of ~ 1.0 J/cm², surface roughening is measurable in both types of tungsten. While the powder met tungsten starts out twice as rough as the single crystal material, the latter roughens significantly after 30000 pulses. It is noteworthy that the single crystal material does not appear to roughen until a certain number of pulses are exceeded (somewhere between 10⁴ and 3×10^4 pulses). At a high fluence of ~ 1.2 J/cm², catastrophic damage is done to both types of tungsten. This is not a surprise as calculations predict that the melt temperature would be reached in a single pulse. This condition significantly exceeds that which is expected in an IFE power plant.

XAPPER will be equipped with a fast, non-contact optical thermometer, which has been developed and built at the University of California at San Diego. This instrument will be installed in December 2004, and it will provide a ~ 1 ns resolution in surface temperature measurements. The thermometer will provide a much desired confirmation of fluence measurements and modeling predictions.

Additional work is being done to more accurately measure and predict peak X-ray fluences. A three-axis motor-driven manipulation system for the ellipsoidal condensing optic has been installed. Testing regarding repeatability of optic positioning and fluence measurements is underway.

Considerable additional work is needed. Since a XAPPER fluence of 0.8 J/cm² would replicate the expected peak surface temperature for the IFE armor, exposures at this fluence are desired. Longterm (>10⁵ pulses) exposures at fluences of 0.7 J/cm² should be completed to determine if the tungsten will eventually roughen at this marginally lower fluence. Additional exposures at 1.0 J/cm^2 are needed both to replicate the original exposures (recall that the 1.0 J/cm^2 was an approximate value based partially on anecdotal evidence) and to learn if the roughening continues with an increasing number of pulses. It is unclear if roughening, unless severe, poses a significant risk to an IFE armor. If the roughening observed at $\sim 1.0 \text{ J/cm}^2$ were to continue in a linear fashion, the surface roughness would be nearly 1 mm after 1 year of operation at 10 Hz. Would this be a problem? Of course, the roughening could continue to grow linearly until some point at which a critical crack size was exceeded and then a large chunk could break off of the armor. These questions only can be answered with addition X-ray exposures.

Coordination of our activities with those of the other HAPL experiments is needed. Specifically, it is desirable to have each facility run a series of experiments that are as closely matched as possible. This coordination effort is underway and future irradiations will be conducted starting at the same temperature and rising to the same peak surface temperature for each experiment. Later investigations will include matching of the peak stresses as well.

Additionally, swapping of exposed samples between the experiments will be conducted in the future. For example, it would be interesting to see if a previously X-ray irradiated powder met tungsten responds in the same manner to ion implantation as does a virgin sample.

Ultimately, it may be of interest to operate XAP-PER with different gases. Argon produces an X-ray spectrum ranging from 250 to 300 eV, while nitrogen yields 400 to 500 eV photons. These higherenergy photons would be more penetrating, and thus, would more accurately mimic the expected IFE armor conditions. Finally, operating XAPPER at a higher repetition rate also might be of interest. With an inter-pulse dwell time of as little as 100 µs, the thermal transient fully dissipates between pulses. This easily supports a repetition rate of 1 kHz, which would enable completion of 10^8 pulses in a 28 h run. Exposing candidate armor materials to their expected lifetime of pulses might be a very interesting endeavor. Discussions with the X-ray source developer, PLEX LLC, are underway.

References

- M.R. Zaghloul, M.S. Tillack, T.K. Mau, Laser-induced damage of metal mirrors under long-term exposure at shallow angle of incidence, in: 19th SOFE, Atlantic City, NJ, 21–25 January 2002.
- [2] L.J. Perkins, personal communication, 2003.
- [3] Target spectra for reference targets can be found at: <<u>http://</u> aries.ucsd.edu/ARIES/WDOCS/ARIES-IFE/SPECTRA>.
- [4] A.R. Raffray, Chamber threats, design limits and design windows, J. Nucl. Mater., these proceedings doi:10.1016/ j.jnucmat.2005.08.015.
- [5] R.R. Peterson, D.A. Haynes, I.E. Golovkin, G.A. Moses, Phys. Plasmas 9 (5) (2002) 2287.
- [6] J. Blanchard, C. Martin, Thermomechanical effects in a laser IFE first wall, J. Nucl. Mater., these proceedings doi:10.1016/j.jnucmat.2005.08.007.

- [7] M. McGeoch, Radio-frequency-preionized xenon Z-pinch source for extreme ultraviolet lithography, Appl. Opt. 37 (1998) 1651.
- [8] J.F. Latkowski, R.P. Abbott, R.C. Schmitt, Pulsed X-ray exposures and modeling for tungsten as an IFE first wall material, Fusion Sci. Technol. 47 (3) (2005) 591.
- [9] Temperature-dependent properties can be found at: ">http://www-ferp.ucsd.edu/LIB/PROPS/>.
- [10] A.R. Raffray, personal communication, November, 2004.
- [11] X-ray opacity data has been taken from the Center for X-ray Optics at: <<u>http://www.cxro.lbl.gov/optical_constants/filter2.</u> html>.