

# Laser inertial fusion dry-wall materials response to pulsed ions at power-plant level fluences <sup>☆</sup>

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## Abstract

Pulses of MeV-level ions with fluences of up to 20 J/cm<sup>2</sup> can be expected to impinge on the first-wall of future laser-driven Inertial Fusion Energy (IFE) power plants. To simulate the effect of these ions, we have exposed candidate dry-wall materials to ion pulses from RHEPP-1, located at Sandia National Laboratories. Various forms of tungsten and tungsten alloy were exposed to up to 1000 pulses, with some samples heated to 600 °C. Thresholds for roughening and material removal, and evolution of surface morphology were measured and compared with code predictions for materials response. Tungsten is observed to undergo surface roughening and subsurface crack formation that evolves over hundreds of pulses, and which can occur both below and above the melt threshold. Heating and Re-alloying mitigate, but do not eliminate, these apparently thermomechanically-caused effects. Use of a 3-D geometry, and/or use of the tungsten in thin-film form may offer improved survivability compared to bulk tungsten.

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## 1. Introduction

The first-wall of an Inertial Fusion Energy (IFE) power plant will be subjected to intense pulsed neutrons, X-rays, and energetic ions produced at the reactor chamber center. An experimental investigation is underway to test IFE chamber wall materials response to X-rays on the Z facility, and to ions on Repetitive High Energy Pulsed Power (RHEPP-1). Both Z and RHEPP-1 are located at Sandia National Laboratories. Materials exposure results on Z are discussed elsewhere [1]. We report here on repeated exposure of first-wall armor materials to intense ion beams. The RHEPP-1

accelerator was used to produce fluences of either helium or nitrogen ions of up to 10 J/cm<sup>2</sup> per shot (for up to 1000 shots per sample), with ion energies as high as 1.8 MeV (for doubly charged nitrogen). The ion current pulsewidth is 200–500 ns at the treatment location, which is shorter than the debris ion arrival time, but comparable to the fusion product arrival time expected in future reactors. Accordingly, the fluence thresholds discussed below may be lower than what might be expected for an actual reactor.

A laser IFE reactor may be operated at a pulse rate as high as 10 Hz. This amounts to an annual total of 3 × 10<sup>8</sup> pulses to the first wall. Besides the considerable thermal energy delivered to the wall surface on each pulse, there likely will be effects of a thermomechanical nature caused by the pulsed energy delivery, such as expansion of the near-surface region against the (relatively) unheated subsurface, fatigue responses, etc. Ions with up to 20 J/cm<sup>2</sup> fluences and several MeV energies will impinge normal to and penetrate well below the surface (1–10 μm). Surface sputtering is secondary to ablation/sublimation of the wall surface. To ensure

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survivability, effectively no erosion of the flat wall surface per pulse can be tolerated (<1 nm). In addition to high melting/vaporization points, additional durability can be expected from materials with good ductility, fatigue response, and high thermal conductivity. ‘Engineered’ materials, e.g., with non-flat geometry, may also be constructed in a way to better distribute the heat load and mechanical stresses.

The work reported here is a continuation of experiments previously detailed [1]. In the earlier study, the roughening threshold for powder metallurgy (PM) W was measured at 1.25 J/cm<sup>2</sup> using the RHEPP-1 N beam. The threshold for W alloyed with 25% Re (W25Re) was found to be significantly higher, above 3 J/cm<sup>2</sup>. This is evidently due to the higher ductility of the alloy. Total exposure in this previous study was limited to 75 pulses.

We discuss here the results of exposure of tungsten in various forms, and compare with other metals such as Mo. For this paper, samples were subjected to as many as 1000 pulses. In addition, samples of W and Mo (both PM) were heated to 600 °C, a temperature which is above the brittle–ductile transition temperature (DBTT) for both elements. The increased number of exposures has led to new conclusions about the surface response of most of the materials tested, compared to the previous work. This will be detailed below.

Flat-geometry samples of pure tungsten (W) formed by both powder metallurgy and CVD were prepared, along with single-crystal W. Flat samples of W25Re were also prepared. This alloy is used in applications such as thermocouple wiring, where increased ductility is desired. The W25Re melting point is about 400 K less than that for pure W (3680 K). In addition, graphite in the form of ‘velvet’, i.e., long fibers oriented parallel to the flux direction, was coated with a sputtered layer of 1.6 μm-thick W, and exposed to ion doses. Measurements of surface roughening and ablation were made with 1 and 2-dimensional profilometry. Effects on surface topology and near-surface microstructure were studied using secondary electron microscopy (SEM) and cross-sectional transmission electron microscopy (XTEM). Surface composition was studied with energy dispersive spectrometry (EDS). In the discussions below, these results are compared with predictions of materials response from the BUCKY [2] and SIM [3] modeling codes.

## 2. Experimental setup and test procedure

Materials were exposed to ion beam fluences on the Repetitive High Energy Pulsed Power (RHEPP-1) facility, which is described elsewhere [4,5]. The ion beam is formed in the MAP (Magnetically confined Anode Plasma) ion diode. For this work, beams were formed

from either nitrogen (45% N<sup>2+</sup>/45% N<sup>1+</sup>/10% H) or helium (~100% He<sup>+</sup>). The He pulse may better simulate the effect of the fusion products arrival, and features a longer pulsewidth than N (approaching 0.5 μs compared to 200–300 ns). Fluence is limited to about 3 J/cm<sup>2</sup> compared to the 8 J/cm<sup>2</sup> possible with N. Diode voltage varied from 600–900 kV over the 100 ns power pulse with nitrogen, and was somewhat lower with He (~500–600 kV). The diode–sample distance was typically 40 cm. Current densities varied, depending upon sample location, from 200 A/cm<sup>2</sup> peak to less than 20 A/cm<sup>2</sup> over a 150 cm<sup>2</sup> total area. Nearby charge collectors monitored the ion beam fluence as a function of sample location. The ions penetrate several microns into the test material, and induce a rapid heating, followed by rapid cooling (10<sup>9</sup> K/sec) as the thermal pulse diffuses into the substrate. At high enough fluences, sublimation/ablation of the surface occurs. Each pulse delivers roughly a monolayer of ions (~10<sup>14</sup>/cm<sup>2</sup>) at depths up to ~1 μm, so that compositional changes in the treated material due to the added ions can be said to be minor. With 1000 pulse exposures, some effects due to ion implantation might be possible, although a sample of W exposed to 1000 nitrogen pulses and examined with XTEM showed no signs of gas accumulation.

## 3. Ion beam exposure experiments

The response of materials irradiated by a high-power ion beam can be quite complex. Surface roughness, as measured by profilometry, can be increased or decreased [6], depending upon the untreated topology. Roughening can be caused by a number of mechanisms, including alloying with volatile elements such as sulfur, the presence of entrained gases, and the material grain size and microstructure. Surface morphology evolution can thus be expected to be a function of grain size and near-surface microstructure, as will be seen in the discussion of PM, single-crystal, and CVD forms of tungsten below. Roughening can also differ, for the same fluence, depending upon the ion composition of the beam.

The roughening threshold for PM W was previously measured at 1.25 J/cm<sup>2</sup> using the RHEPP-1 N beam. The sample (unheated) was exposed to over 50 pulses during this measurement. To determine if the PM W surface would remain unaffected by the ion beam below this threshold, a polished and unheated sample was subjected to 400 pulses of the MAP N beam, over a fluence range per pulse between 1 and 3.7 J/cm<sup>2</sup>. Surface roughness ( $R_a$ ) was then measured with 1-D profilometry. The results are plotted in Fig. 1, along with several representative points for untreated PM W, and a single point for treated PM Mo. (The untreated values are plotted at a fluence only for representational purposes.) Note that below the 1.25 J/cm<sup>2</sup> value, the W surface

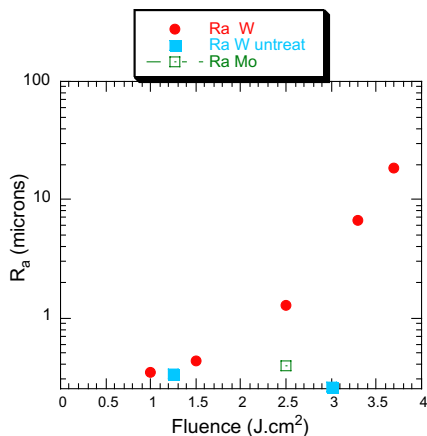


Fig. 1. Surface roughness ( $R_a$ ) of PM W (unheated), as a function of N beam fluence, after 400 pulses. Data points for untreated W and PM Mo also included.

roughness does not increase above the untreated level. Above this value, however, the  $R_a$  rapidly increases with dose, reaching over  $20 \mu\text{m}$  at  $3.7 \text{ J/cm}^2$  fluence, with Peak-to-Valley (P–V) surface excursions exceeding  $70 \mu\text{m}$ . Even at  $2.5 \text{ J/cm}^2$ , the W surface roughness significantly exceeds that of the PM Mo sample exposed.

A set of samples was prepared for a 450-pulse exposure to the MAP He beam. Three forms of W – PM, CVD, and single-crystal, were exposed at a fluence of  $1\text{--}1.25 \text{ J/cm}^2$  per pulse, i.e., at or below the roughening threshold measured for the N beam. Also included were polished samples of W25Re, and pure Re. The latter were exposed at fluences up to  $2.5 \text{ J/cm}^2$ . The W samples were heated to  $600 \text{ }^\circ\text{C}$ , whereas the W25Re and Re were treated at room temperature. After 450 pulses, the most notable result is that the PM W exhibited a relatively high level of roughening, compared to both the W25Re and Re, and even compared to the CVD and single-crystal W. Evidently, the roughening threshold is lower when He ions are used for treatment, compared to N ions. This is consistent with anecdotal experience that heavier ions produce smoother surface topology, compared to that produced by H or He ions on RHEPP-1. No other samples in this shot series, other than the PM W, showed significant roughening beyond the as-polished levels. SIM modeling predicts the He beam fluence needed to melt the W (heated to  $600 \text{ }^\circ\text{C}$ ) at  $2.3 \text{ J/cm}^2$ , and melt fluences for the W25Re and Re at about  $2.7 \text{ J/cm}^2$ . Given the typical 20% fluctuation in fluence between pulses, both the heated W and unheated W25Re and Re samples were exposed to fluences that could have caused their surfaces to melt, if only briefly.

Fig. 2(a) and (b) are photomicrographs (5X magnification) showing surface relief from the PM and single-crystal W samples, respectively. (The length scale bar

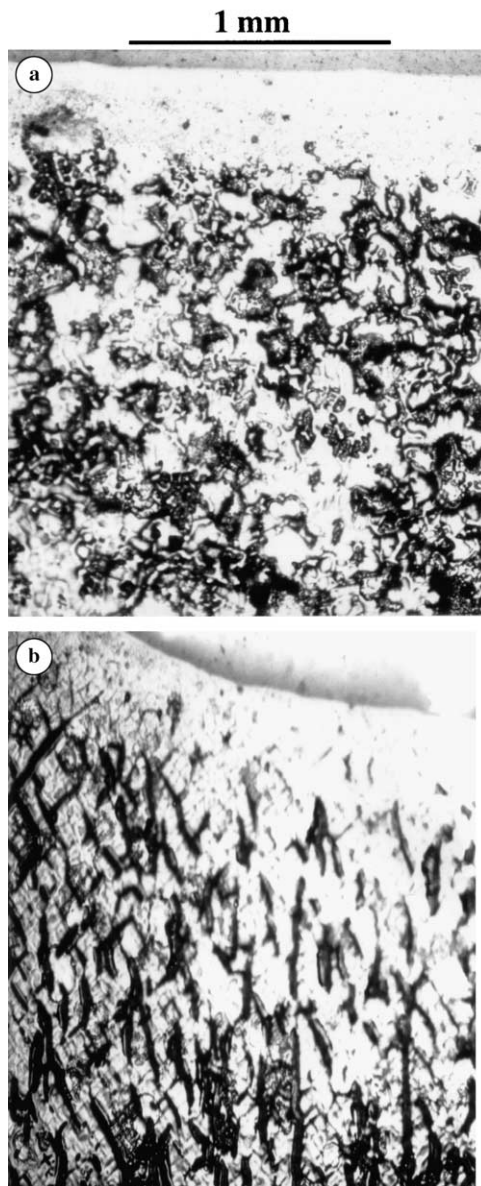


Fig. 2. (a) Micrograph (5X magnification) of PM W surface after 450 He pulses at  $1\text{--}1.25 \text{ J/cm}^2$ . Surface  $R_a$  measured at  $2\text{--}3 \mu\text{m}$ . Treatment interface is at top. (b) Micrograph (5X magnification) of single-crystal W surface after 450 He pulses at  $1\text{--}1.25 \text{ J/cm}^2$ . Surface  $R_a$  measured at  $0.5\text{--}0.6 \mu\text{m}$ .

shown in Fig. 2(a) applies also to Fig. 2(b).) The relief features on the single-crystal surface show an alignment, presumably along crystallographic planes, whereas the PM surface looks more chaotic. The  $R_a$  of the single-crystal W ( $0.5\text{--}0.6 \mu\text{m}$ ) is significantly less than that of the PM W ( $2\text{--}3 \mu\text{m}$ ). However, the  $R_a$  of the W25Re remained typically below  $0.2 \mu\text{m}$ , even at  $2.5 \text{ J/cm}^2$ , without heating to  $600 \text{ }^\circ\text{C}$ . This indicates that the pres-

ence of the 25% Re is leading to a reduction of the roughening behavior with repeated He beam exposure.

When compared with the prior study [1], the 400-pulse exposure results shown in Fig. 1, and the 450-pulse He series suggest that surface relief can take many pulses to evolve. To investigate the materials response of a number of metals, a 1000-pulse series was undertaken with the MAP N beam. Samples were exposed of: copper, aluminum 1100, titanium grade 2 (commercially pure), W25Re, Re, and W and Mo in PM form. The W and Mo were heated to 600 °C. The 1000 pulses were taken in groups of 200, i.e., after every 200 pulses, the samples were demounted,  $R_a$  and P–V were measured with 1-D profilometry (DekTak), and the samples re-mounted, until 1000 pulses were attained. The radius of the DekTak tip was 12.5  $\mu\text{m}$ . The P–V data are shown in Fig. 3, for a fixed fluence per shot of 2.5  $\text{J}/\text{cm}^2$ . The melting point of course varies for the various metals for this fixed fluence, but in each case the metal is exposed to near or above melt. The Al 1100 sample almost reaches its vaporization point at the surface for this fluence. The data points for each metal are shown connected, to better clarify the trends with pulse number. The surface roughness of the PM W is observed to increase greatly after the first 200 pulses, from  $<5$  to  $>35$   $\mu\text{m}$  from 200 to 600 pulses, after which it appears to stabilize. The surface roughness of several of the metals, notably PM Mo and Ti-2, while initially much lower than the W, increases steadily for the entire 1000 pulses, with no signs

of saturation. The rate of roughness increase for the W25Re and Re is much lower, but also shows a steady upward trend, whereas the Cu surface roughness hardly changes over the entire series. After the series was completed, SEM observations were made of each surface. These observations also included other fluence exposures than those shown in Fig. 3, notably 4  $\text{J}/\text{cm}^2$ . The surface topology of each metal is observed to undergo an evolution that is different in each case. The general appearance is that shown of the PM W surface in Fig. 2(a), i.e., a periodically chaotic pattern. The scale length of surface features, and the evolution of this scale length with fluence, and with pulse number, can be different for each exposed metal. This is presumably due to the different mix of surface melting and vaporization points, elastic modulus, ductility, etc., in each case.

The practical implications of this behavior trend are unclear. While 1000 intense ion beam pulses might be thought of as considerable in a research setting, in the lifetime of a future IFE reactor, this amount of pulses can be reached in less than 2 min. The W roughening appears to reach a saturation level after about 600 pulses. The Mo and (unheated) W25Re surfaces show significantly less roughening over the 1000 pulses than the PM W, but their  $R_a$  shows no signs of saturation, and could continue to grow beyond the 1000 pulses examined here. In addition, the P–V plot in Fig. 3 apparently understates the actual surface relief evolution. Two-dimensional profilometry (Veeco) observations were made of each of the surfaces above. In the case of the W and Re surface, very deep and narrow cracks were observed, narrow enough so that the 12.5  $\mu\text{m}$  DekTak tip was not able to detect them. The depth of these cracks is considerable, tens to perhaps hundreds of micrometers in extent. The exact amount is unknown, because the tracking capability of the Veeco instrument was exceeded. The origin of these cracks is not known at this point, but may be related to fatigue cracking. Since the materials investigated here are assumed to be applied in a sheet form (perhaps 200–300  $\mu\text{m}$ ) over a structural substrate (such as ferritic steel), the presence of such deep-lying cracks suggests that if fatigue is the cause, then the cracks may be expected to reach the depth of the structural material underneath [7]. Since the roughening threshold for the heated PM W occurs considerably below its melting temperature, we infer that the dominant cause of the surface roughening and crack formation is not melt/solidification, but instead is probably thermomechanical stress. That is, the heated near-surface layer is forced to expand and contract against the unheated W substrate, and the resulting strain cracks the surface.

A BUCKY simulation was performed of the W sample (unheated) irradiated by the He beam, at a fluence of 1.3  $\text{J}/\text{cm}^2$ , i.e., at the roughening threshold for W measured for the MAP N beam. For this simulation,

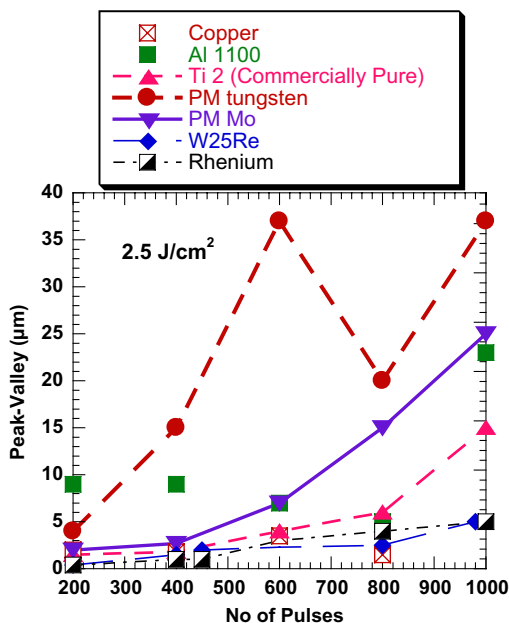


Fig. 3. Evolution of Peak–Valley excursions for a number of metals, as a function of number of MAP N ion pulses. Fluence is 2.5  $\text{J}/\text{cm}^2$ .

temperature-dependent thermal conductivity and heat capacity were used, as well as temperature dependent thermo-mechanical data for tungsten from the ITER materials handbook. This data include yield strength, elastic modulus, coefficient of expansion, and Poisson's ratio.

The result of the simulation is shown in Fig. 4. Note that at this fluence, when the peak surface temperature is 1800 K, i.e., well below the W melting point, the W may yield at a depth as deep as 3  $\mu\text{m}$ . This is the result of a single He pulse. For repeated exposures, the effects of such yielding, i.e., cracks and surface relief, could be expected to be cumulative.

Previously [1], samples of carbon composite were exposed to ion pulses, and the surface roughness measured. The carbon fibers in the composite survived the exposure much better than the matrix material between the fibers. In the limit as the matrix material is removed, and the fibers arranged lengthwise along the beam direction, one has a 'velvet' or 'brush' geometry. The tips of the fibers in this configuration would be exposed to the full effect of the ion beam, but the length of the fibers (millimeter length scale) constitute a 'get lost' region, where ions contact the side walls of the fibers at glancing angle, and thus deliver a greatly reduced fluence. If the fibers could then be coated with an armored material that stops incoming fusion products before they enter the graphite, then tritium buildup in the graphite could be avoided.

A velvet consisting of carbon fibers was coated with 1.6  $\mu\text{m}$  of sputtered W thin film. The tops of the fibers received the full sputtered thickness, and the side walls received correspondingly less. The fiber coverage was 2%, i.e., only 2% of the carbon substrate onto which the

fibers were attached was covered by fibers. The velvet was exposed to 200 pulses of the MAP N beam, at fluences as high as 6  $\text{J}/\text{cm}^2$ . This fluence is well above the carbon sublimation fluence, and is roughly equal to the W vaporization threshold for the N beam. Visual inspection of the velvet indicated no damage evident, even at the highest fluence. The surface appeared more black than the untreated velvet, presumably because any fibers oriented non-vertically were removed by the beam, thus decreasing the macroscopic optical reflection. The carbon substrate was undamaged. Closer inspection with SEM revealed that at lower fluences, the W coating appeared to be mechanically removed, and appeared as debris at the bottom of the fibers where they attach to the substrate. At the higher fluences, the fiber tip response can be divided into two general types. Where the tip remained flat, the W was removed, and the W appeared as a small bead attached to the side wall of the fiber. On about 10–15% of the fibers, however, the tip geometry appeared sharpened, coming to a point. On these fibers, the W still remained. That this could occur with an originally 1.6  $\mu\text{m}$ -thick sputtered W layer is remarkable. It may be possible to heat-treat the fibers to increase the adhesion tendency of the W, and also to sharpen all the fiber tips to recreate the geometry that seemed to retain the W at the tip. Such a W-coated carbon velvet may then become a viable engineered surface for long-term survivability.

#### 4. Summary

Various forms of tungsten, including CVD, single-crystal, and PM, as well as alloyed with Re, were exposed to pulsed ions on RHEPP-1. Dose levels and spectra are similar to those expected in future laser IFE direct-drive reactors. W-coated carbon 'velvet' was also exposed. The PM form of tungsten appears to undergo thermomechanical roughening, starting at doses lower than needed to melt the surface. The evolution of this roughening occurs over hundreds of pulses. In addition, there appears to be deep-lying cracks that may be fatigue-caused, that may extend tens to hundreds of microns below the surface. Roughening is higher for equivalent fluences of He ions, as compared with N ions. Heating to 600  $^{\circ}\text{C}$  reduces this tendency to some extent. BUCKY simulations show that the tungsten yields to a depth of several microns for even modest fluences.

Use of single-crystal or CVD tungsten is not practical for a reactor wall. However, what these studies suggest is that some modification of powder metallurgy tungsten appears necessary for its use in a pulsed reactor environment. Use of tungsten in thin-film form, as with the W-coated carbon velvet, may also prove to be a way to avoid the deleterious properties of W in bulk form.

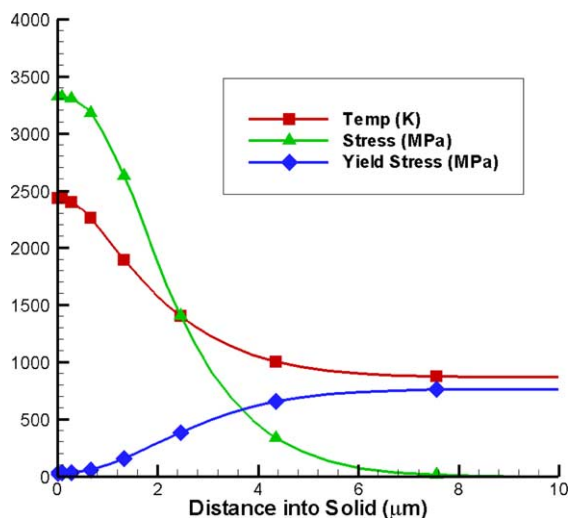


Fig. 4. BUCKY simulation of PM W, heated to 600  $^{\circ}\text{C}$ , treated with single pulse of MAP He beam at 1.3  $\text{J}/\text{cm}^2$ .

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