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DEVELOPMENT AND EVALUATION OF FIRST WALL MATERIALS
FOR THE NATIONAL IGNITION FACILITY
6/12/96

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ABSTRACT

Several low-Z refractory materials are evaluated for use as the NIF first wall in terms of their cost and ability to survive laser light, target emissions and debris, as well as be cleanable and not outgas excessively. The best performers contain B, C, or both, with B₄C being the best overall. It appears possible at this time that plasma-sprayed B₄C can be fabricated with low enough porosity and cost to be preferred to hot-pressed B₄C, the conservative choice.

I. INTRODUCTION

The NIF target-chamber first-wall material must survive high x-ray, stray light, debris, and shrapnel exposures without generating vapors or particulates that will excessively contaminate the final optics debris shields or other components in the target chamber, thereby making them susceptible to subsequent damage [1]. It also must not outgas excessively during chamber pumpdown [2] or activate significantly under high neutron fluxes [3]. Furthermore, it must be cleanable by CO₂ pellets or some other method to remove target debris and tritium [4]. Finally, the first wall must be developed and fabricated for a reasonable cost. These requirements provide a challenge to material science.

It was decided early that the first wall would consist of removable panels fastened to the inside of the chamber. The panels could be made completely from the first-wall material or by coating or bonding first-wall material to aluminum panels. Sprayed coatings would be less expensive than thick panels. For example, B₄C coatings are estimated to cost about \$1M less than 6-mm-thick hot-pressed panels.

II. MATERIALS TESTED

Existing data and preliminary modeling were used to select a group of materials for more extensive evaluation [5]. One of the primary criteria is a large x-ray deposition

depth, which indicates a low-Z material. At the same time, the material should have a high melting or boiling point to minimize ablation. These criteria limited the list of potential candidates to various forms of B, B₄C, C, Al₂O₃, MgAl₂O₄, SiC, Si₃N₄, and SiO₂. Be was not considered seriously to minimize possible environmental, health, and safety problems.

Potential first-wall materials were acquired from both DOE and commercial suppliers. Oak Ridge (Y-12) supplied 10 plasma-sprayed coatings of B₄C, B, Al₂O₃, MgAl₂O₄, and mixtures thereof on aluminum panels [6]. Additional plasma-sprayed B₄C and B coating plus solid panels of C, B₄C, s, SiC, Si₃N₄, SiO₂ and Al₂O₃, B₄C composites (Norbide and a cermet) were acquired from ten companies. Coatings were typically 200-500 μm in thickness. One important property of plasma-sprayed material is porosity, as will be shown later. Polished cross sections of four plasma-sprayed B₄C samples in the test program are shown in Figure 1.

III. TEST PROGRAM

To test the survivability of these materials to NIF-like conditions, they were subjected to a variety of physical tests, including impact testing at Y-12, CO₂ abrasion at Y-12 and LLNL, x-ray exposures at AWE (Helen) in England, CEA (Phebus) in France, and LLNL (Nova), 351 and 1053 nm light exposure at LLNL, and vacuum outgassing at LLNL. The tests at other facilities provided preliminary guidance, and only the LLNL tests will be discussed in any detail here.

A. Material Removal by X-rays

The primary threat to the first wall is x-ray radiation. Results will be described only briefly because they are described in detail elsewhere [5]. A range of removal amounts occurring by various mechanisms were observed for different materials. For reference, 0.1 μm of material removal from the 10-m-diam. chamber corresponds to 30 cm³, a potentially troublesome amount for the final optics.

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Figure 1. Polished cross sections of four plasma-sprayed B_4C samples used in first-wall development studies. Sample (a) was sprayed in vacuum and has the lowest porosity, outgassing, and erosion rate during CO_2 cleaning. Sample (b) tended to crumble during polishing and has the most erosion during CO_2 cleaning. Samples (c) and (d) are intermediate in porosity and performance.

As expected from theory, B, B_4C , C compounds performed the best under x-ray exposure. SiC performed nearly as well. All suffered roughening at 220 eV blackbody fluences greater than $1.5 J/cm^2$ due to thermal crazing or other mechanisms, which made accurate material removal measurements difficult. Even so, we estimate less than $0.01 \mu m$ of B and B_4C would be removed for a 20 MJ ignition yield (equivalent to $1.85 J/cm^2$ on Nova), nearly all as particulates, and only slightly more for C and SiC. Solid B and B_4C appear untouched by x-ray fluences of $1 J/cm^2$, but plasma-sprayed B and B_4C show evidence of superficial melting. An important, not-yet-understood observation is that hot-pressed B_4C from one company is much more prone to thermal crazing than B_4C from another company.

SiO_2 performed only slightly worse, having a removal depth of about $0.02 \mu m$ for fluences equivalent to a 20 MJ yield NIF shot. However, it was removed by vaporization rather than particulate ejection. Alumina of various forms performed worse than SiO_2 , apparently because liquid splash and what appears to be a thermal crazing/solid spall mechanism. Si_3N_4 performed significantly worse, because it thermally decomposes to molecular nitrogen, which aids the vaporization of the Si. Aluminum was the worst of all materials tested, having nearly $1 \mu m$ removed at a 20-MJ-yield fluence by a combination of vaporization and liquid splash.

B. Material Damage by Laser Light

The first wall may be exposed to as much $0.6 J/cm^2$ of

3ω ($0.35 \mu m$) scattered light over 3 ns from direct drive experiments and lower amounts of 1ω ($1.06 \mu m$) and 3ω for other experiments. Further, it would be desirable to use the first wall directly as a beam dump for unconverted light (1ω over 20 ns) if the beam could be appropriately spread out. Consequently, polished samples of most of the same materials were exposed in a vacuum to 1 and 3ω light at various fluences and pulse lengths to characterize their damage thresholds and mechanisms. Damage observed by laser light was similar to that observed for 200-250 eV blackbody x-rays. In followup experiments at 3ω , material removed from selected, unpolished (more realistic) materials was collected on the laser entrance optic.

For a 2.4 ns pulse of 3ω light, visible surface damage starts for full-density B, B_4C , and SiC at about $0.5 J/cm^2$. The damage is faint thermal crazing for B and slight mottling for B_4C and SiC. Photographs of B_4C damage are shown in Figure 2. The thermal crazing becomes severe for B at about $1 J/cm^2$. In contrast, B_4C shows more severe mottling at that fluence, and thermal crazing begins about $1.5 J/cm^2$. SiC shows more pronounced mottling and enhanced original grain boundaries as fluences increases over the same range, but clear thermal crazing was not observed. Plasma-sprayed B_4C with a porosity of 10% or so showed reflectivity changes associated with superficial surface melting at $0.25 J/cm^2$. The reflectivity changes at higher fluence were always greater than for solid B_4C , but no thermal crazing was observed. C shows similar reflectivity changes and enhancement of visible grains, but no clear evidence for material removal by profilometry. The damage threshold of full-density Al_2O_3

Figure 2. Surface damage to hot-pressed B₄C from one manufacturer caused by exposure to 0.35 μm laser light.

(spalling of microscopic grains) exceeds 1.2 J/cm²; more spalling is observed at equivalent fluence from HVOF (high velocity oxygen fuel) sprayed Al₂O₃. Increasing pulse length increases damage thresholds slightly. Increasing from 2.4 to 6 ns, equivalent damage occurred at about 20% higher fluence for B, B₄C, and SiC and about 50% higher fluence for Al₂O₃.

Damage thresholds at 1ω are higher than for 3ω because of the lower absorption coefficients. For B and B₄C, the thresholds increased threefold. Some mottling and isolated crazing was observed for B at 3 J/cm² and 6 ns, but significant crazing was not observed until 3.5 J/cm² for a 6 ns pulse and 4.7 J/cm² for a 58 ns pulse. Initial damage for hot-pressed B₄C was observed at 2.5, 3.5, and 5.0 J/cm² for 2.4, 6, and 58 ns pulses, respectively, and substantial mottling and crazing was observed about 1 J/cm² higher. Plasma-sprayed B₄C showed reflectivity changes and surface melting at only 0.8 J/cm² for 2.4 ns. SiC received barely perceptible mottling at 1 J/cm² for both 2.4 and 6 ns. As fluence increased to 3.5 J/cm², about 10% of the surface was covered by microscopic pits, and the majority of the surface was pitted to a depth of a couple μm at 5 J/cm². C showed reflectivity changes and enhanced grain contrast at 0.8 J/cm² and 2.5 ns, but any ablation was less than 0.2 μm at 4 J/cm². Spalling of both HVOF and hot-pressed Al₂O₃ starts about 1.8 J/cm² and 2.4 ns. By 4.5 J/cm², the HVOF coating ejects 10+ μm of material, and hot-pressed Al₂O₃ has pits 5-10 μm deep over 25% of its surface. For 58 ns pulses, Al₂O₃ has insignificant damage at 4 J/cm² and less than 25% surface pitting at 8 J/cm².

Laser-ablated material from hot-pressed and plasma-sprayed B₄C, hot-pressed and HVOF Al₂O₃, SiC, and C was collected on a facing optic about 4 mm away. The optic was rotated 90° after each of 4 shots to monitor weathering effects. All materials received a 0.6 J/cm² shot in a first round, and either 1 (all but Al₂O₃) or 2 J/cm² of 3ω light. For all materials except C, the first shot tended to "clean" the target, covering several percent of the collecting optic with particles <5 μm in diam. Few

particulates were generated from any material by the fourth shot at 0.5 J/cm², and only HVOF Al₂O₃ at 2 J/cm² generated particulates on the fourth shot of the higher fluence. The collected particulates looked fairly angular, suggesting they arrive as solids, except for Al₂O₃ at the high fluence, which looked like molten splats. Two materials, SiC and C, also formed brown thin films over the beam area, presumably due to evaporation of target material. The spots were fainter on the first shot but fairly uniform thereafter. White light transmittance was about 95% and 75% for C and about 90% and 75% for SiC exposed to 0.6 and 1.0 J/cm², respectively.

B₄C (pressed or sprayed) and hot-pressed Al₂O₃ perform well in terms of the stray light threat, primarily a concern for direct drive. At the lower fluence, surface damage is negligible and particulate generation seems to be a startup transient that may be minimized by appropriate pretreatment. SiC and C appear less desirable because the stray-light threat is close to the threshold for evaporation and subsequent deposition of an absorbing thin film. In terms of using the first wall as a beam dump, primarily for 1ω light, B₄C and Al₂O₃ are comparable and can tolerate long pulses of about 4 J/cm² without damage.

C. Damage of Contaminated Optics

Since no material tested guarantees zero contamination of the final laser optic for 20-MJ ignition shots, the propensity of fugitive first-wall material to damage optics is also important. Consequently, damage thresholds were measured for optics contaminated with material ablated by laser irradiation of B₄C and Al₂O₃ and by x-ray irradiation of SiO₂. It was postulated that SiO₂ and Al₂O₃ might be substantially less damaging to the optics because they are transparent.

Preliminary experiments using optics from the preceding section indicated that the difference between B₄C and Al₂O₃ is far less than anticipated. Particulates of both were blown off by the first laser pulse of 5-10 J/cm², and catastrophic damage did not occur for 10-20 additional

Figure 3. Sequential cleaning and ultimate damage of an SiO_2 optic contaminated with B_4C particulates.

shots. An example for B_4C is shown in Figure 3. Damage was about the same for comparable levels of B_4C and Al_2O_3 contamination. Since Al_2O_3 particulates are much more likely to be formed by x-ray exposure and since neither material has a significant advantage for laser exposure, B_4C is clearly the better of the two. Consequently, additional experiments using B_4C and higher quality optics were conducted.

A sample of as-pressed (not ground) B_4C was first cleaned by bombardment with CO_2 pellets and preconditioned in vacuum with four 3ω laser shots of fluence 1.8, 1.2, 1.8, and 0.6 J/cm^2 . Material mobilized by 3 ns shots of 0.6 and 1.4 J/cm^2 was then collected on optics stationed 50 cm from the target. Next, material mobilized by a 3 ns shot of 1.4 J/cm^2 was collected on an optic stationed at 5 cm. The optics had initial damage thresholds (600 shots of 3 ns 3ω light with no damage) of 15 J/cm^2 .

The mildest exposure (0.6 J/cm^2 , 50 cm) caused the 600-shot damage threshold to decrease to about 10 J/cm^2 , but the optic survived 100 shots at 13 J/cm^2 before major damage. The optic contaminated at 50 cm with increased particulates from the 1.4 J/cm^2 shot still had a 600 shot damage threshold of about 10 J/cm^2 , but damage occurred within 3 shots at 13 J/cm^2 . The center of the 2-cm optic had a visible brown spot in the center, presumably from vaporization of B_4C . This region had an optical density of 0.1, corresponding to a thickness of 6 nm, and a damage threshold of less than 1.5 J/cm^2 . The annular region outside the brown region had moderate damage after 100 shots of 6 J/cm^2 .

Fused SiO_2 has relatively low ablation by x-rays and would be a viable first-wall candidate if the vaporized silica from the first wall deposited on the debris shield in a high-damage-threshold thin film. This was explored briefly by exposing one optic to 3.5 J/cm^2 on Nova and collecting the ablated material on another optic. The collecting optic had substantially increased scattering centers in darkfield microscopy, and the damage threshold of the collecting optic dropped from 10 to 3 J/cm^2 .

Though not as controlled as desirable, this experiments shows that deposited SiO_2 films are unlikely to have high damage thresholds due to incorporation of particulates, oxygen-deficient stoichiometry, or both. SiO_2 is not clearly superior to B_4C in terms of optical threat to the debris shield and is inferior in other respects.

The implication of these results is that B_4C is not a greater threat to the final optics assembly than optically transparent materials such as Al_2O_3 and SiO_2 . The laser-cleaning of B_4C particulates from optics allows 5-20 lower fluence shots prior to replacement of the debris shield for the next high-yield shot. Not only does this reduce the frequency of debris shield replacement, it also reduces occupational exposure to radiation from chamber materials activated by the high yield shot by allowing time for the activation to decay.

D. Outgassing Measurements

The NIF target chamber must reach 5×10^{-5} Torr in less than 2 hours after exposure to air. Preliminary design of the vacuum system indicates that the first wall material can have an outgassing rate of $1 \times 10^{-6} \text{ T-L/s-cm}^2$ after 1 hour of high vacuum pumping. Further, the pressure chamber must reach 1×10^{-6} Torr in a few more hours for cryogenic experiments, thereby requiring an ultimate outgassing rate of 2×10^{-8} or lower.

Outgassing rates of single 15-cm-square or multiple 10 -cm-square panels were measured in the LLNL Vapor Processing Laboratory by an orifice pressure differential method. All materials were baked in a vacuum oven for 24 hours then exposed to room humidity for 8 hours. One material, low-porosity plasma-sprayed B_4C , was tested with two orifice sizes with equivalent results. The same material was also tested after venting to air passed through a 1 ppm humidity filter.

A summary of outgassing results is shown in Figure 4. A couple of conclusions are obvious immediately. Hot-pressed B_4C panels do not outgas significantly and qualify

Figure 4. Outgassing rates for candidate first-wall materials. High porosity B₄C represents data for materials in Figures 1b&c. Low porosity B₄C represents the material in Figure 1a. Dry vent B₄C represents a re-test of the low porosity B₄C after venting to 1 ppm humidity air.

easily, as long as a suitable bonding agent to the backing plate can be found. High-porosity plasma-sprayed B₄C coatings do not qualify. B₄C coatings with porosities of 5-6% appear to meet the two hour specification, and with proper control of the vent gas humidity, can probably reach the cryogenic pressure requirement within a reasonable time.

E. CO₂ Cleaning Experiments

Solid panels were superior performers compared to thermal spray coatings in preliminary CO₂ cleaning experiments. Initial harsh CO₂ cleaning showed the potential for stripping entire coatings, while hot-pressed B₄C was essentially unphased [4]. The challenge was to develop both coating and appropriate test procedures which would qualify those coatings as surviving appropriate cleaning conditions.

The most sensitive measure of material removal from these rough materials turned out to be gravimetry. Early results showed a larger mass removal on the first cleaning, presumably removal of surface contamination and loose material, followed by a steady decrease in mass per cleaning at a rate dependent on the material porosity. One plasma-sprayed B₄C coating, the same one that passed the vacuum outgassing specification, approached the performance of hot-pressed B₄C under cleaning conditions that remove the majority of vapor-deposited Au and Cu from hot-pressed B₄C. A 2.5x5-cm piece lost <1 mg after 3 cleanings, which corresponds to <0.1 μ m per cleaning.

Another piece received 100 cleanings (several decades of NIF) with acceptable erosion.

Early experiments tested removal of vapor-deposited Au and Cu. Cleaning conditions that remove >90% of gold films 50-250 Å from hot-pressed B₄C remove only 75% of same from plasma-sprayed B₄C (sample 1a). The same conditions remove three-fourths of 250 Å Cu films on hot-pressed but only about half from plasma-sprayed B₄C (sample 1a). Only a third of thin Cu films (<150 Å) are cleaned from hot-pressed B₄C. On average, 60 Å of Cu remains after cleaning from all thicknesses.

F. Lifecycle Testing

In the complicated and somewhat uncertain NIF-chamber environment, interactions between metal deposits, x-ray exposures, light, and shrapnel may interact to change performance of the first wall material relative to that expected from investigations of threats individually. Consequently, 2.5x5-cm pieces of potential B₄C materials underwent cyclic exposures to Nova target emissions and CO₂ cleaning to assess relative long-term performance. Materials were placed at 40 or 80 cm from chamber center for one or two weeks at a time (total of 5 weeks). At 40 cm, they received 20 to 25 exposures per week to a maximum x-ray fluence of 1 J/cm², roughly equivalent to a 5 MJ ignition shot.

Figure 5 shows the change in weight for eight potential first-wall materials: two hot-pressed B₄C materials (different suppliers), two B₄C-rich composite materials (Norbide and a B₄C-Al cermet), and four plasma-sprayed B₄C samples (three different suppliers). The weight change upon Nova exposure depends on both the amount of metal and semi-volatile organics picked up in the chamber, and the weight loss upon cleaning includes both metal contamination and coupon material.

The high-purity B₄C panels (Ceradyne and Cercom) performed significantly better than the B₄C-composite materials. While there may be a less expensive form of B₄C panels with satisfactory performance, the excellent performance of one of the sprayed coatings makes such a panel less interesting. The sprayed coatings performed in order of their porosity, as seen by comparing the weight losses to the porosities estimated from Figure 1. Erosion by CO₂ cleaning appears to be reduced to the level required for the estimated porosity of 5-6% present in B₄C 1a.

Results for the efficiency of metal removal by CO₂ cleaning are less encouraging. For cleaning conditions in which vapor-deposited films are mostly removed from hot-pressed B₄C, most of the Nova deposits remain on the

Figure 5. Net weight changes in eight candidate first wall materials. Weight loss from plasma-sprayed B₄C increases with porosity. The two hot-pressed and the lowest porosity (1a) plasma-sprayed materials perform very well. A weight loss of 1 mg corresponds to 0.3 μ m of B₄C erosion.

coupons, except for the high-porosity coatings, which clean by erosion of the B₄C matrix. Perhaps the metal deposits are more tightly bonded to the B₄C by chemical reactions at the interface enhanced by the high temperatures of the multiple x-ray exposures. Whether the cleaning efficiency can be improved by increased severity while maintaining acceptable loss of first-wall material remains to be demonstrated.

IV. CONCLUSIONS

B₄C appears to be the best material for the NIF target-chamber first wall. It is readily available commercially in a variety of forms; it has the least material removal by x-rays and has a high threshold for vaporization; it tolerates laser exposure to the levels required and may even be suitable for using large areas of the first wall as a beam dump; and its particulates tend to be cleaned from optics upon exposure to relevant laser pulses, thereby allowing 5 to 20 lower fluence experiments to follow a high-yield ignition shot without changing the debris shields.

While hot-pressed B₄C panels were the initial conservative choice for the NIF first wall, recent and substantial progress in low-porosity plasma-sprayed B₄C coatings suggests that sprayed coatings on Al panels are a viable option. Lifecycle material removal from cyclic exposures to x-ray, metal, and light on Nova and to CO₂

cleaning is comparable for the best hot-pressed materials and the best plasma-sprayed coatings. The best sprayed material also meets or is close to meeting all outgassing specifications. However, some additional materials studies and fabrication of full-scale panels will be required before a final decision can be made.

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