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EVALUATION OF B₄C AS AN ABLATOR MATERIAL FOR NIF CAPSULES

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ABSTRACT

Boron carbide (B₄C) is examined as a potential fuel container and ablator for implosion capsules on the National Ignition Facility (NIF). A capsule of pure B₄C encasing a layer of solid DT implodes stably and ignites with anticipated NIF x-ray drives, producing 18 MJ of energy. Thin films of B₄C were found to be resistant to oxidation and modestly transmitting in the infrared (IR), possibly enabling IR fuel characterization and enhancement for thin permeation barriers but not for full-thickness capsules. Polystyrene mandrels 0.5 mm in diameter were successfully coated with 0.15–2.0 μm of B₄C. Thicknesses estimated from optical density agreed well with those measured by scanning electron microscopy (SEM). The B₄C microstructure was columnar but finer than for Be made at the same conditions. B₄C is a very strong material, with a fiber tensile strength capable of holding NIF fill pressures at room temperature, but it is also very brittle, and microscopic flaws or grain structure may limit the noncryogenic fill pressure. Argon (Ar) permeation rates were measured for a few capsules that had been further coated with 5 μm of plasma polymer. The B₄C coatings tended to crack under tensile load. Some shells filled more slowly than they leaked, suggesting that the cracks open and close under opposite pressure loading. As observed earlier for Ti coatings, 0.15- μm layers of B₄C had better gas retention properties than 2- μm layers, possibly because of fewer cracks. Permeation and fill strength issues for capsules with a full ablator thickness of B₄C are unresolved.

I. INTRODUCTION

NIF capsule design depends on a balance among what will properly implode and what can be properly built,

fielded, and diagnosed. All capsule designs will be made primarily from low Z materials (C or lower atomic number) with a few percent of oxygen or nitrogen allowable and a few percent of a higher Z material (e.g., Br, Ge, or Cu) added for opacity control. The most commonly considered materials are organic polymers and Be, but a review of potential materials in mid-1995 suggested that boron and its carbides should be considered. Subsequent calculations indicated that undoped B₄C has very favorable implosion attributes. In addition, it is relatively easy to sputter coat. As a result, we have explored some of the fabrication and fielding issues associated with B₄C capsules, including optical properties and permeability.

II. DESIGN ISSUES

One- and two-dimensional LASNEX calculations predict that a capsule using pure B₄C ablator material encasing a layer of solid DT can be ignited successfully using anticipated NIF laser pulses resulting in an x-ray drive with a peak blackbody-equivalent temperature of 300 eV. One such design is shown in Fig. 1. This capsule achieves ignition and significant fuel burn, producing 15 MJ of energy. The thickness of the ablator is greater than for designs of plastic capsules and comparable to that in Be designs. B₄C is able to both shield the DT fuel from preheat effects and develop an ablation-front density profile favorable to implosion stability without having to include a higher Z dopant, as is necessary with CH/Br or Be/Cu capsules. Preliminary designs with straight boron did require a dopant at 300 eV, so given that fabrication would likely be more difficult, this option was not pursued. However, B₄C does have a disadvantage relative to doped Be in that a design was not

Figure 1. Specifications for a B₄C NIF ignition capsule yielding 18 MJ when driven with 300-eV peak-temperature x rays. The absorbed energy is 199 kJ, and the fuel ρ is 1.75 g/cm².

found that would successfully ignite using only a 250-eV hohlraum temperature drive (gotten either from a lower laser energy or an increased hohlraum size). Further details are given elsewhere.¹

III. FIELDING ISSUES

The physical properties of B₄C influence potential characterization and fielding strategies, which in turn affect fabrication goals. B₄C is a very strong material, with a fiber tensile strength (S_t) of 2–6 GPa.^{2,3} Since a capsule can hold a pressure of $2S_t \rho r$, a 1-mm-radius capsule with a 0.15-mm-thick wall with appropriate grain structure could theoretically contain pressures in the GPa range. However, B₄C is also very brittle, and microscopic flaws or grain structure will likely limit the fill to a factor of ten or so less than the fiber values. What pressure could be ultimately contained by a sputtered B₄C capsule is an open research issue. Further, the hydrogen permeability is unknown, so a filling scenario is still an open question. B₄C is very stable thermally, so high-temperature diffusion may be possible as long as any deposition mandrel is also thermally stable or removable by some means (e.g., diffusion out or uniform reaction with an inner B layer). Finally, volumetric heating of the DT ice layer by radioactive decay provides a driving force in the presence of external cooling to symmetrize and smooth the DT ice layer.⁴ However, any unplanned steady-state temperature gradients within the hohlraum would cause long-scale steady-state ice-layer nonuniformities if not smoothed by the capsule wall, and the ability of the capsule to accomplish this smoothing is proportional to its cryogenic thermal conductivity. Further, it would be desirable and

perhaps necessary to characterize and maybe enhance the DT ice layer smoothness. The IR opacity⁵ and cryogenic electrical conductivity⁶ of the B₄C affect the ability to accomplish these goals.

A. Optical Density

Both B and B₄C are semiconductors with a band-gap in the near infrared region. Pure crystalline boron has a minimum in optical absorbance of about 5 cm⁻¹ for 3 μ m light,⁷⁻⁹ the appropriate wavelength for DT layer enhancement by IR heating, and the absorption coefficient changes little when cooled from room temperature to 150K.¹⁰ A 0.15-mm-thick boron capsule wall would absorb less than 8% of the incident light, although it would also reflect about 30%.

However, the literature is inadequate for accurately assessing the optical properties of sputtered B or B₄C, so films of various thicknesses were magnetron sputtered onto silica or KBr substrates. Films of a few to a couple hundred nm are progressively darker brown in optical transmission. Average optical densities (reflection plus absorbance) of B₄C measured with a Sargent–Welch model DT63 densitometer were 0.18, 0.28, 0.32, and 0.36 for thicknesses of 14, 29, 33, and 66 nm. The optical densities did not change over a 6-month period, indicating that negligible oxidation occurs for even the thinnest films. Results for B were similar, but the thicker films wrinkled due to compressive stress during deposition.

Optical densities of sputtered films were also measured as a function of wavelength from the IR to ultraviolet (UV, Fig. 2). The absorbance falls off exponentially from the UV to the IR, except for the thickest film, where the development of an interference fringe is evident (Fig. 3). (Similar results were obtained for boron.) 50% transmittance through 14-, 33-, and 66-nm-thick films occurs at 420, 730, and 1020 nm, respectively. Data from 328 to 2622 nm were fitted to two dispersion theories: one derived for amorphous semiconductors with parabolic bands,¹¹ and the standard damped harmonic oscillator. Slightly better fits were obtained with the damped harmonic oscillator function:

$$= (n + ik)^2 = 1 + \frac{A}{\left(\frac{\omega^2}{\omega_0^2} - 1\right) - i\gamma},$$

where $\gamma = 2\omega_0 c / \hbar A$, giving $\hbar \omega_0 = 6.68$ eV, $\hbar A = 2.77$ eV, and $\hbar \gamma = 6.69$ eV (7.9, 3.76, and 9.65 for B).

Figure 2. Optical densities of thin B_4C films sputtered onto glass substrates.

A 2- μm film transmits more than half of 2- to 8- μm IR light. A 0.34-mm slice of commercial hot-pressed B_4C was found to be opaque in the IR, but it contained detectable amounts of graphite by x-ray diffraction, so it may be more absorbing than a comparable thickness of high-purity sputtered B_4C . Even so, these results indicate that thin coatings of B_4C used as permeation barriers would allow optical characterization and possibly enhancement of DT ice layers, but full-thickness B_4C capsules probably could not use optical methods. A by-product of this effort was the development of a calibration of optical density versus thickness that was useful for monitoring the thickness and uniformity of thin B_4C coatings on capsules.

B. Electrical and Thermal Conductivity

The cryogenic thermal and electrical conductivities affect the ability to form a uniform DT ice layer by beta-layering and enhance its smoothness, respectively. The cryogenic thermal conductivity of both boron and boron carbide are reviewed by Türkes *et al.*¹² The conductivity at 20K of various B_4C samples varied from 0.02 to 0.10 $Wcm^{-1}K^{-1}$. While this is substantially below the value of about 30 $Wcm^{-1}K^{-1}$ for single crystalline Be, it is within a factor of ten of commercial Be containing 2% Mg,¹³ which is more what one might expect for a capsule doped with Cu for opacity control. However, sputtered B_4C will likely be amorphous or nearly so. At 20K, -B is near its maximum conductivity

Figure 3. Optical density of a thicker, free-standing, sputtered B_4C film in the visible and infrared.

of 4 $Wcm^{-1}K^{-1}$, but amorphous B has a conductivity of only about 0.01 $Wcm^{-1}K^{-1}$. Even though the conductivity of a B_4C capsule has considerable uncertainty, it is undoubtedly still greater than plastic, which has a value of about 0.0015 $Wcm^{-1}K^{-1}$. Consequently, B_4C capsules could use a cryogenic hohlraum with considerably looser thermal-gradient specifications than required for plastic capsules.

While the excellent implosion stability of both Be and B_4C capsules likely means that the native DT ice roughness is probably adequate, the low electrical conductivity of $10^{-8} \text{ }^{-1}m^{-1}$ of pure B at 100K initially suggested that smoothing by rf heating might be possible. The conductivity would have to be lower than the DT gas value of $7 \times 10^{-6} \text{ }^{-1}m^{-1}$ for the method to be viable. However, various authors¹⁴⁻¹⁶ report DC conductivities of 3×10^{-3} to $> 100 \text{ }^{-1}m^{-1}$ for B_4C at 73K, presumably dependent on impurities and variations in B/C ratio. Kormann and Zuppiroli¹⁷ report the electrical conductivity of B_4C as a function of both temperature and frequency. While their DC conductivity of $6 \times 10^{-6} \text{ }^{-1}m^{-1}$ is within the range of possible utility, the conductivity at low temperature increases linearly with frequency on a log-log scale. At 20K and 1 GHz, as appropriate for NIF capsules, we estimate a conductivity of about 0.05 $\text{ }^{-1}m^{-1}$ from their graphs. Consequently, DT ice layer smoothing by rf heating does not seem to be viable for B_4C capsules. Neither is it likely for boron capsules because of the need for high-Z doping for opacity control.

Figure 4. Optical micrograph (transmitted light) of two 450-μm-diameter polystyrene microshells coated with nominally 1500 Å of B₄C. The difference in darkness corresponds to different thicknesses due to an initial flash coating before the shells were bounced. One of the shells has picked up dust particles because of static charge, but the other shell appears to remain clean.

Figure 5. Optical micrograph (reflected light) of a 450-μm-diameter polystyrene microshell coated with about 2 μm of B₄C showing defects caused by sticking during the coating process. Other shells showed wrinkling or buckling from heat. Later coating runs had fewer of these defects, but the sticking problem was not eliminated consistently.

IV. SPUTTER-COATING OF POLYSTYRENE MANDRELS

Standard Nova polystyrene mandrels (450-μm diameter by 3-μm thick) were sputter-coated with B₄C using three magnetron sputter guns and a piezoelectric-driven bounce pan. Significant agglomeration problems were encountered during some of the runs, but six coating runs with thicknesses ranging from about 50 nm to 2 μm were completed. The deposition rate averaged 0.3 μm per hour. The coated shells tended to charge with static electricity, picking up dust and occasionally jumping out of their dish. Even so, it was possible to handle the shells sufficiently for characterization. It was easily possible by optical density under the microscope to estimate the coating thickness and coating uniformity of the B₄C, based on an optical density calibration curve derived from flat films. Some of the thin coatings were clearly nonuniform when a flash coating was applied prior to start of the bounce pan. Both dust and coating nonuniformity are evident in Fig. 4.

Figure 5 shows optical micrographs of the thickest B₄C capsules showing surface defects caused by

sticking and possible thermal deformation by plasma heating. This is not yet a serious concern, because relatively few runs were attempted and the optimal coating conditions have not yet been found. Figure 6 shows two fracture cross sections and the surface of the coated shells, by SEM. Some defect growth cones were found, and most appeared to start during the coating run rather than at the polystyrene surface. This may have been caused by particulate ejection from the sputter gun, which could be remedied through higher quality and better preconditioned sputter targets. Figure 6 also compares a fractured B₄C capsule wall with the same magnification of a Be capsule wall fabricated under comparable conditions. The finer diameter and straighter columnar growth in the B₄C are clearly evident. While this structure would give a more uniform material for implosions and possibly a smoother surface, it may also be harder to fill and weaker in lateral tension than the unique twisted microstructure of the Be formed under these conditions.

The ultimate objective for maximum strength would be to form an amorphous structure with no columnar growth. The microshell coatings produced in this work have a slightly more pronounced columnar structure than reported earlier,^{18, 19} but they are probably still either

Figure 6. Scanning electron micrographs. Left side: two side views of fractured B_4C coatings, one 1.6 μm thick and the other 2.0 μm thick, on polystyrene microshells. Right bottom: top view of the B_4C surface finish. Right top: fracture surface for a Be capsule. The B_4C has a much finer columnar structure than Be.

amorphous or nanocrystalline. X-ray diffraction, SEM, and transmission electron microscopy of the earlier sputtered B and B_4C films uncovered no columnar growth for thicknesses less than 1 μm and no crystallinity for thicknesses up to 6.8 μm . More work is needed to achieve this type of film on capsules.

V. PERMEABILITY MEASUREMENTS

The permeability of the coated shells was measured by diffusion filling with Ar and monitoring the leakage by x-ray-induced fluorescence. Argon was used for convenience at this early stage. Even though the correlation between DT and Ar permeabilities observed for plastic capsules²⁰ is not quantitatively accurate for

B_4C , Ar does give a qualitative indication of the DT permeability of the films.

Three capsules each from four batches (12 capsules total) with nominal thicknesses of 0.15 μm (two runs), 0.5 μm , and 2 μm were first coated with about 5 μm of plasma-polymerized trans-2-butene. Some of the shells were cracked after this coating, although it is possible that very fine cracks were present but not observed during the less rigorous examination prior to coating. Two uncracked shells with a 0.15- μm coating attained very little initial Ar fill and had half-lives too long to measure (> 1 week). The average leakage half-lives for the cracked shells were 0.7 hours for the 0.5- μm B_4C coating and about 10 hours for the 0.15- μm B_4C coatings. Three shells of 2 μm thickness, for which

cracks were not observed because they were optically opaque, had an average Ar half-life of 4.1 hours. A possible explanation for the longer gas retention of the 0.15- μm coating is that it has fewer cracks. A similar phenomenon of more cracks and a higher permeability for a thicker film has been observed for 50- and 100-Å Ti coatings over poly-*m*-methyl styrene.²¹

During these experiments, it was noticed that the measured Ar fill pressure was lower at initial time than expected from the Ar leak rate. Consequently, a second set of fill experiments was conducted, and the time constant for filling was estimated from the increase in Ar signal. Filling half-lives ranged from 23 to 134 hours for both 0.15- μm and 2- μm B₄C coatings; these are 3 to 50 times longer than observed for leakage. After this second fill, the leakage was measured a second time, yielding half-lives equal to or up to twice as long as the original half-lives. Therefore, the permeation appears to be dominated by cracks, and permeability increases and decreases as the cracks open and close under internal or external pressure.

Microscopic examination suggested that the B₄C cracked, while the polymer layers did not. Consequently, a few alternating very thin layers of B₄C and polymer could substantially reduce permeability by increasing the effective diffusion length through the polymer as long as any cracks in the B₄C layers do not coincide. This approach would be particularly effective if a relatively impermeable polymer such as polyimide were used between the B₄C layers. The B₄C would probably perform best for thicknesses of a few tens of nm or less, and this approach would solve both the nonuniformity and radiation sensitivity of the current PVA coating. B₄C would also be an option for a permeation barrier embedded within a Be capsule wall, if the permeability of Be itself cannot be properly tailored by deposition conditions.

VI. CONCLUSIONS

B₄C provides a possible backup to Be as a more stable implosion alternative to an all-plastic NIF capsule. Undoped, its implosion characteristics rival the best doped Be designs at 300 eV, although it performs less well at lower drive temperatures. With only a modest effort, it was possible to coat polystyrene microshell mandrels with thin coatings of B₄C. The sputtered B₄C has a finer microstructure than sputtered Be, and thin films are quite impermeable to Ar from room temperature to 140°C when not cracked. This may lead to smoother and less permeable capsules than for Be. However, it may also make the NIF capsules more difficult to diffusion

fill. Evaluation of this issue requires thicker shells, but they are more difficult to make with B₄C than Be because of the 3 to 7 times slower sputtering rate. B₄C does have the advantage that thin coatings, which are most relevant for gas-filled noncryogenic shots on either Nova, Omega, or NIF, are partially transparent, so coating thickness and uniformity are readily measurable by optical means. The infrared transmittance is much higher, but it is doubtful that enough IR light could pass through a NIF-thickness shell to be useful for either fuel layer diagnostics or layer enhancement heating. Also, the GHz-range, cryogenic electrical conductivity is too high to use rf heating of free electrons in the DT gas to enhance DT ice smoothness. How aggressively B₄C capsules should be pursued in the future depends primarily on whether any problems arise for meeting the evolving specifications for the doped-Be NIF ignition targets.

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