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Subject: Space Shielding Materials for Prometheus Application

Enclosure: Shield Materials Information

Dear Sir:

This letter provides a summary of the Naval Reactors Prime Contractor Team (NRPCT) research into the use of various shield materials for a Prometheus application. Most of the information provided herein, including categorization of materials as "primary, alternate, or eliminated," was developed in support of a planned down-selection to allow focused materials development. The down-selection recommendation was not completed due to Prometheus program restructuring.

SUMMARY:
At the time of Prometheus program restructuring, shield material and design screening efforts had progressed to the point where a down-selection from approximately eighty-eight materials to a set of five "primary" materials was in process. The primary materials were beryllium (Be), boron carbide (B4C), tungsten (W), lithium hydride (LiH), and water (H2O). The primary materials were judged to be sufficient to design a Prometheus shield—excluding structural and insulating materials, that had not been studied in detail. The foremost preconceptual shield concepts included: (1) a Be/B4C/W/LiH shield; (2) a Be/B4C/W shield; (3) and a Be/B4C/H2O shield. Since the shield design and materials studies were still preliminary, alternative materials (e.g., 7B or 10B metal) were still being sorely considered, but at a low level of effort.

Two competing low mass neutron shielding materials are included in the primary materials due to significant materials uncertainties in both. For LiH, irradiation-induced swelling was the key issue, whereas for H2O, containment corrosion without active chemistry control was key. Although detailed design studies are required to accurately estimate the mass of shields based on either hydrogenous material, both are expected to be similar in mass, and lower mass than virtually any alternative. Unlike Be, W, and B4C, which are not expected to have restrictive temperature limits, shield temperature limits and design accommodations are likely to be needed for either LiH or H2O. The NRPCT focused efforts on understanding swelling of LiH, and observed, from approximately fifty prior irradiation tests, that either casting or thorough outgassing should reduce swelling. A potential contributor to LiH swelling appears to be LiOH contamination due to exposure to humid air, that can be eliminated by careful processing. To better understand LiH irradiation performance and mitigate the risks in LiH development for a project with an aggressive schedule like JIMO, some background or advanced development effort for LiH should be considered for future space reactor projects.

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BACKGROUND:
The information contained herein was obtained through literature review and engineering analysis (a significant amount being performed by Oak Ridge National Laboratory (ORNL)). The down-selection of primary and alternate shield materials was based largely on ORNL shield design scoping studies (References (a), (b), (c)) in conjunction with literature review of promising materials. ORNL varied candidate materials in a self-optimizing shield design code, then compared final mass values for the shield. This was a crude but effective way to identify promising materials. Overall, about eighty-eight materials were screened.

A detailed review of the material properties available in the literature for the primary materials Be and LiH was completed (Reference (d)). Further, a detailed historical review of LiH irradiation-induced swelling was completed (Reference (e)), including new quantum mechanical modeling, a hypothesis for reducing LiH swelling, and an approach to testing the hypothesis. Lithium hydride has been judged the best neutron shielding material in prior US low-mass reactor shield efforts (Aircraft Nuclear Propulsion [ANP] Program, SNAP, SP-100) and was flown on SNAP-10A, and ~30 Russian space reactors (primarily TOPAZ systems). However, as discussed herein, it was not clear LiH should be used in a Prometheus application.

DISCUSSION:
In order to efficiently utilize resources and meet challenging delivery schedules, a shield materials down-selection was planned, despite missing key information about some of the candidate materials. At the time of project restructuring, the NRPTC was in the process of preparing a formal recommendation to down-select to five primary materials: Be, B4C, W, LiH, and H2O. A limited effort to identify and evaluate promising alternative materials (e.g., 11B or 10B) was also planned. The enclosure provides summary level information on the rationale behind selecting the primary and alternate shielding materials, and a general justification behind eliminating a select set of other candidate materials (generally, those materials that were of the most interest, but not chosen for continued evaluation).

Down-selection criteria were largely based on four factors: (1) ORNL shield mass estimates using candidate materials; (2) judgment of the required additional irradiated materials testing to qualify a material; (3) cost and deliverability; (4) design complexity issues (e.g., very narrow material temperature limits or the requirement for a containment vessel or other support system). Other risk issues, e.g., safety concerns with handling a material, were also factored into the selection. In the cases of Be, B4C and W, there were no significant issues noted. In fact, the Be/B4C/W shield concept was favored because it appeared to be low risk. If it is ultimately found to be mass-competitive with LiH or H2O containing shields, it would likely be selected.

Given the need to minimize mass, LiH and H2O were selected as primary materials despite the need for irradiated materials testing, as well as other design complexities and risks. A summary of the Prometheus shield design efforts is provided in Reference (f).

**Gamma Shielding Materials**
**Primary Gamma Shielding Materials:**
Tungsten was selected as a primary gamma shielding material. Tungsten has well known shielding properties, is readily available, manufacturable, low toxicity, and relatively inexpensive. Tungsten is commonly used in irradiation testing as a gamma heating material, with no material degradation noted. It was not expected to require any materials development. A complete literature search on this material should be performed to ensure there are no gaps in the material properties that warrant testing.

**Alternate Gamma Shielding Materials:**
None selected.

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Eliminated Gamma Shielding Materials:
On a gamma attenuation-per-mass basis, a higher performing alternative to tungsten is depleted uranium (U). However, all U-bearing materials were eliminated due to high expected costs and handling difficulties relative to W, and the likelihood that the U would transmute into plutonium, that could subsequently fission and cause higher doses aft of the shield. Other materials that were eliminated include YH3, ZrHx, and ZrB2 each of which provide combined neutron and gamma attenuation benefits, but were not shown to provide shield mass benefits to justify their development. Zirconium carbide was also eliminated based on shield mass screening studies.

Neutron Shielding Materials
Primary Neutron Shielding Materials:
Beryllium provides some gamma attenuation as well as neutron moderation but no neutron absorption. A Be slab at the front of the shield may also be beneficial as a mounting plate for the aeroshell, potentially as a housing for neutron detectors, and to remove heat. Beryllium is a commonly used reflector material, with extensive irradiated materials data in the literature. From a manufacturing standpoint, large, high quality Be slabs up to ~114cm diameter are commercially available.

Boron carbide is a high temperature material that provides some gamma attenuation with low capture gamma production, as well as neutron moderation and excellent thermal neutron absorption. Boron carbide is a commonly used neutron poison, especially in light water reactor control rods. As such, it has been irradiation tested extensively. High quality plate is commercially available and relatively inexpensive, but fabrication issues were not fully investigated.

Lithium hydride provides excellent neutron moderation and absorption. ORNL shield material screening studies identified that all minimum mass shields included LiH. However, irradiation-induced swelling was identified as a jugular issue for LiH. The NRPCT completed a literature review and analysis (including quantum mechanical modeling) of LiH, focused on swelling (Reference (e)). It was observed that either casting or out-gassing the LiH may reduce swelling. The NRPCT hypothesized that LiOH contamination contributes to the swelling, however, further unirradiated and irradiated materials testing and engineering evaluation are required to determine whether this hypothesis is correct. If so, this jugular issue may be resolvable.

Pure H2O is an excellent neutron moderator, but without the addition of a neutron poison (e.g., dissolved boric acid and/or lithium hydroxide) neutrons would be absorbed by H atoms releasing a gamma ray, which reduces shield effectiveness. Therefore, the NRPCT and ORNL evaluated neutron-poisoned H2O, that is routinely used in operating reactors to control reactivity and in safety systems to ensure reactor shutdown. The differences between these systems and the space reactor shield environment must be evaluated in detail to determine the viability of water for use in the shield design space. For instance, no chemistry monitoring or corrections are envisioned for the spaceship. A jugular issue was corrosion of the containment system over the ~20 year life. Effort should focus on defining an overall system that provides the desired neutron attenuation characteristics, and has the lowest likelihood of having significant corrosion concerns. Radiolytic decomposition of the water is also a concern. As with LiH, irradiated materials/system testing would likely be required.

Alternate Neutron Shielding Materials:
Although there are significant uncertainties with both LiH and neutron-poisoned H2O, no back-up hydrogenous material was recommended. Should both LiH and neutron poisoned H2O be eliminated from consideration, an all-Be/B4C/W (or perhaps a solid 10B) shield would probably

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meet the design requirements.

Boron was identified as a potential combined gamma and neutron absorption material, but detailed studies of boron material properties were not completed, and preliminary shield mass studies did not show a mass advantage relative to primary material based shields. Advantages of an all-B shield are simplicity, both in thermal design and manufacturing, and low toxicity relative to Be and LiH.

Eliminated Neutron Shielding Materials:
Organic materials (primarily phenyl-based, high temperature oils, saturated hydrocarbon polymers (e.g., polyethylene), and high temperature capable, high hydrogen density polyisobutylene (PIB)) were studied in some depth, including select ORNL mass screening studies. As a class, organics irreversibly decompose due to radiolysis evolving significant amounts of hydrogen gas, which likely requires engineering accommodations (e.g., pressure relief system). Temperature limits on high hydrogen density organics, e.g., polyethylene, were very restrictive, e.g., < 400K. Polyisobutylene showed promise as a high temperature, high hydrogen density organic, but irradiated material testing was likely required to qualify this material. Given that ORNL studies did not show mass savings for organics, and the unavoidable hydrogen gas loss, this class of materials was eliminated.

Carboranes (e.g., B10C2H12) were also studied, but eliminated due to only being available in limited quantities at high cost.

CONCLUSIONS:
Five primary materials, Be, B2C, W, LiH and neutron-poisoned H2O were chosen for focused development/qualification to support the three shield design concepts being considered prior to program restructuring:

1. Be/B4C/W/LiH
2. Be/B4C/W
3. Be/B4C/H2O

Little development or testing was expected to be needed for Be, B2C, or W. Lithium Hydride was identified by ORNL screening studies as a required neutron shield material for a minimum mass shield. Since LiH has the jugular issue of irradiation-induced swelling, which may not be resolvable, neutron-poisoned H2O was also included as a primary material. However, H2O containment corrosion in a radiolytic environment over the twenty year Prometheus mission was also considered a jugular issue. Both the LiH and H2O issues are expected to require irradiated materials and/or systems testing to be resolved.

Structural and other materials (e.g., insulation) were not studied prior to program restructuring. Due to the shield material studies being preliminary, further screening of alternate materials (e.g., 10B or 11B metal) was planned.

CONCURRENCES AND ACKNOWLEDGEMENTS:
The Manager, Space Materials (Simonson), the Manager, SPP Space Power Plants Systems (Schwartzman), the Manager, Space Plant Materials (Ohlinger) and the Manager, FSO-Shield Design Development (Collins) concur with this letter.

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NR ACTION REQUESTED:
This submittal is for information. No NR action is requested.

Very truly yours,

Rose Lewis, Engineer
Fuel and Shield Technologies

Brian Campbell, Manager
Fuel and Shield Technologies

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REFERENCES:


(c) ORNL/LTR/NR-JIMO/05-27, "Reactor Shield Optimization in Support of the Prometheus 1 Project Preconceptual Design Analysis," dated October 2005.


(e) NRPCT letter MDO-723-0048 "The Evaluation of Lithium Hydride for Use in a Space Nuclear Reactor Shield, Including a Historical Perspective, for NR Information," dated December 9, 2005.

(f) NRPCT letter SPP-67210-0011, "Shield Design Summary," To Be Issued.
Enclosure to MDO-723-0049:

Shield Materials Information

Authors:
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James Nash

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The purpose of this enclosure is to present some of the key data from literature searches on candidate shield materials for a Prometheus reactor shield. This information was originally collected and analyzed in support of efforts to down-select materials for focused development. It is intended to aid future researchers studying various reactor shielding material options. Emphasis is placed on key properties and jugular issues, rather than a comprehensive collection of all properties for the candidate materials—the latter is available in Reference (a) for LiH and Be.

Shield design typically involves layering shield materials in specific combinations and sequences to achieve overall goals of gamma and neutron attenuation at a minimum mass. The design must also account for heat transport, thermal limits, structural concerns (e.g., launch and other loads, reactor support, etc), and potentially accommodate penetrations for instrumentation, reflector/drum positioning, and coolant piping. This requires the materials and design to be highly integrated. Both design and shielding material selection studies were performed for a Prometheus application and should be considered complementary to the information included herein (References (b), (c), (d), (e)).

1. SUMMARY

The Naval Reactors Prime Contractor Team (NRPCT) was in the process of down-selecting shield materials to a set of five primary materials, water (H2O), lithium hydride (LiH), beryllium (Be), tungsten (W), and boron carbide (B4C), for development and further evaluation. Boron (10B and 11B) was also going to be evaluated at a low level. The primary materials roughly break down into gamma attenuation material: W, non-hydrogenous neutron and gamma attenuation materials: Be, B4C, and hydrogenous, neutron attenuation materials: LiH, H2O, although each material provides a varying amount of attenuation of both gamma and neutron radiation.

For the non-hydrogenous materials, Be, B4C and W, little materials development was envisioned; however only the literature review of Be was completed in depth. An alternative to the aforementioned materials was solid B, which was interesting from a simplicity and toxicity standpoint, but issues such as cost, fabricability and irradiated material properties remained to be studied.

For the hydrogenous materials, LiH was studied in the most depth, but no testing was performed prior to program restructuring. The most significant issue with LiH use is irradiation-induced swelling, which may preclude the use of this material. The literature includes widely varying measurements of LiH volumetric swelling, ranging from 0% to ~25%, making this a jugular issue. Yet despite this key property uncertainty, LiH was used in the SNAP-10A flight shield and numerous TOPAZ reactor shields for short lifetimes (Reference (f)). The NRPCT developed a hypothesis for the swelling1, which may enable the general use of LiH through careful material processing, but due to program restructuring, the testing required to verify the hypothesis could not be performed. The literature also suggests that LiH swelling might be minimized in the 600 to 800K range, but this was clearer for gamma irradiation than mixed neutron/gamma irradiation (actual conditions in the shield). LiH will also need a slight overpressure of H2

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1 Essentially, lithium hydroxide (LiOH) contamination of the LiH is believed to result in excess hydrogen gas forming in the LiH, which subsequently increases the swelling under irradiation (Reference (f)).

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(<<1 atm), and therefore need to be canned for a space application. Issues with canning design, internal structural requirements, temperature control, and H$_2$ retention remain to be addressed.

In the case of H$_2$O, research was only beginning at the time of program restructuring. However, the general view was that the effects of irradiation on water are well characterized in the literature. The materials performance difficulties with using H$_2$O were expected to be in the area of corrosion of the containment system, especially if dissolved poisons were added to the water to enhance neutron absorption and limit neutron absorption in H, which results in gamma emission. The fact that the shield water would be unmonitored, and chemistry not actively controlled for the anticipated Prometheus mission lifetime, nominally twenty years, was a significant concern. Design concerns were also significant with H$_2$O, primarily due to its sensitivity to temperature. In particular, the design would likely need to accommodate or prevent freezing, and ensure temperature transients did not exceed limits due to steam pressure containment concerns.

Integrated shield studies by Oak Ridge National Laboratory (ORNL) (References (b), (c), (d)) consistently indicated that LiH or H$_2$O containing shields are necessary to achieve the targeted attenuation at the lowest mass. However, design details such as containment, single point failure mitigation, temperature control, internal structure, and piping penetrations tend to increase the mass of LiH or H$_2$O shields more than for non-hydrogenous shields. The use of either material likely involves temperature control; however, the LiH is expected to support a wider temperature range depending on design details and material studies. Generally, any need to control temperature is considered undesirable due to the difficulties of performing a thermal design of a space shield (radiative, conductive and convective heat transport; complicated internal energy deposition; gaps that change over the lifetime, etc.).

Additional gamma and neutron shield materials not recommended for use are also briefly discussed herein. Many were eliminated due to extensive research needed for qualification, cost and/or no noted mass benefit.

2. DISCUSSION

This section will focus on the key materials properties and jugular issues for candidate shielding materials

2.1. Primary Shield Materials

2.1.1. Beryllium (Be)

Beryllium metal is known for excellent thermal properties and light weight (theoretical density is 1.85 g/cm$^3$), but it becomes brittle under modest neutron fluences, and is therefore not likely to be relied upon structurally. As a shielding material, it moderates neutrons well, but does not significantly absorb them. Beryllium has been used as a reflector in reactor plants for decades, and has a vast irradiated materials database, which appears to span the design space of a Prometheus type reactor shield (Reference (g)). Brush Wellman sells high quality Be metal, but the material is relatively expensive—in part due to the hazards associated with Be exposure, both in the lungs, and more

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recently identified, as a skin irritant. Beryllium is the nominal first wall (closest to reactor\(^2\)) material for the shield, because it moderates fast neutrons, but also because the aeroshell could use Be in this position to benefit from prior aeroshell qualification efforts. Beryllium is believed to be a likely material to surround neutron detectors in the shield near the reactor, since it would moderate neutrons without significantly absorbing them, and therefore may serve that purpose as well. Beryllium may be used within the shield in areas where the design benefits from increased thermal conductivity (relative to other reactor shield materials), e.g., near piping.

Due to the relatively broad use of Be in reactors (especially reflectors), and indications from the literature that this material has been tested throughout the expected irradiation conditions for a Prometheus application, there were no near term plans to irradiate this material. However, if future researchers identify gaps in the literature (within their design space), irradiation testing should be reconsidered (References (h) - (k)).

No jugular issues were noted for Be.

2.1.2. Boron Carbide (B\(_4\)C)

Boron carbide is a low density (~2.5 g/cm\(^3\)), common nuclear poison due to the high thermal/epithermal neutron absorption cross section of boron, and its high temperature capability (\(T_\text{m} \approx 2723\) K [Reference (l)])). It is relatively inexpensive, and available in different forms, including powders, plate, etc. One commercial application is armor plates. Boron carbide is brittle, and would not be used as part of the shield structure.

The assumption has been that no irradiated materials testing would be required for B\(_4\)C, however, some confirmatory testing may be necessary depending on the form used versus the forms previously tested. Additionally, detailed literature reviews of the material may identify gaps where testing (unirradiated and irradiated) may be required.

No jugular issues were noted for B\(_4\)C.

2.1.3. Tungsten (W)

Tungsten is a very high density (19.3 g/cm\(^3\)) material, but on a per mass basis, it is nearly the best gamma attenuation material, and readily available commercially. Zirconium hydride and uranium compounds also have excellent gamma attenuation capability, but other aspects of these materials complicate their use. Zirconium hydride and depleted uranium are discussed below in separate sections for each material. Tungsten has very high temperature capability, and can be obtained in any reasonable shape needed. The W component of the reactor shield would be optimized to minimize the mass of the shield, while realizing attenuation goals of the shield.

The assumption has been that no irradiated materials testing would be required for W. Future researchers should re-evaluate the need for irradiated (or other) materials testing of W.

No jugular issues were noted for W.

\(^2\) Structural materials will likely be positioned between the Be slab and the reactor, depending on the shield structural design. Shield structural materials were not studied prior to program restructuring.
2.1.4. Lithium Hydride (LiH)

Lithium hydride has a low density (0.775 g/cm$^3$), high neutron absorption cross section ($^6$Li $\sigma_{thermal}$ = 941 barns), high hydrogen content (12.68 wt. %), and a lack of significant secondary radiation upon absorbing a neutron, which in combination makes LiH an excellent neutron shield material. As shown in Figure 1, for a given neutron fluence, LiH provides more neutron attenuation than other candidate materials. Therefore a LiH slab will generally be thinner and have less mass for the same attenuation when compared to the other candidate materials. Other factors that increase the shield mass include structural and canning materials, single point failure mitigation, as well as any other systems, e.g., to maintain LiH temperatures within the required limits.

![Graph showing neutron fluence through monolithic shield materials over thickness and mission time.](image)

**Figure 1: Monolithic Neutron Attenuation (Reference (b))**

Reference (f) compiled and reviewed literature on various material properties of LiH, including its behavior under irradiation. The main concern associated with LiH is irradiation-induced swelling, which varies considerably in the literature. A compilation of the LiH irradiated swelling data reviewed during the Prometheus shield materials studies is shown in Figure 2 and Figure 3 (the source data is also compiled in tabular form in the attachment). The data is plotted by performance as a function of fluence (Figure 2) or temperature (Figure 3), and indicates swelling is minimized in cast or pressed and out-gassed specimens. The maximum expected LiH fluence would be $-1 \times 10^{18}$ n/cm$^2$, and the minimum temperature could be 600K. However, the fluence could be increased and allowable temperature decreased if the swelling issue is resolved. Other trends are difficult to discern given the inconsistency in LiH swelling behavior over a large range of temperatures and fluences. The presence of LiOH as an impurity in a pre-irradiated condition is hypothesized by the NRPCT to be a cause of swelling, but other factors, like microstructural changes in cast LiH may be involved as well. Lithium hydride swelling, possible mechanisms and future irradiation testing strategies are discussed in detail in Reference (f).

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Lithium hydride irradiation-induced swelling is considered a jugular issue. Lithium hydride is a promising, although not fully understood, space reactor shield material, and should continue to be considered as a potential material for future applications. However, sufficient lead time is required to retire both material uncertainties and design risks (thermal and nuclear).

Enriched $^6$LiH was considered, but the mass reduction (~100kg) does not warrant the cost of the enrichment and handling the $^6$LiH as special nuclear material for both testing and construction of the shield. The special handling of $^6$LiH would also increase the schedule risk.

![Irradiation Test Results](image_url)

**Figure 2: Historical Irradiation Test Results from LiH Specimens vs. Fluence**

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2.1.5. Water with Neutron Poisons

Water with neutron poisons may be competitive with other candidate hydrogenous materials at lower temperatures. However, no effort was put forth to study this material.

2.2. Alternative Shield Materials

2.2.1. Boron (B) Metal

Boron metal is a potential alternate shielding material. Boron could be used as the only shielding material in the shield, i.e., the shield would be comprised of slabs of B and necessary structural materials without any other shield materials (Reference (e)). W could be used to reduce mass, depending on radiation limits, structural design, and component shielding available (Reference (e)). Boron has high temperature capability (~1500 K), is a good neutron moderator and absorber, and is relatively low density (~2.35 g/cm$^3$ nat$^{10}$B, ~2.17 g/cm$^3$ 10$^{11}$B). Boron-10 provides a 7.5% mass reduction relative to nat$^{10}$B with a slight improvement in neutron attenuation and no change in gamma attenuation, but at a much higher cost. Boron metal is generally not found commercially, and is not believed to be extensively tested under irradiation conditions, which would necessitate a comprehensive qualification program both in and out of pile.

Table 1 compares a Prometheus shield containing $^{10}$B and Be with three other shield configurations: a Be/B$_2$C shield, a B$_2$C/Be/W/LiH shield, and a B$_2$C/stainless steel/H$_2$O(LiOH) shield (Reference (d)). The Reference (d) comparison is preliminary, and focused on relative mass differences. The mass of the $^{10}$B containing shield is lower
than that of the Be/B₄C shield by ~100 kg, but higher than either the LiH or H₂O based shields (note that canning is not accounted for in the latter cases). It is not clear that the mass differences for the four cases shown in Table 1 are significant given the simplified cases studied. Design details such as containment, single point failure mitigation, temperature control, internal structure, and piped penetrations tend to increase the mass of LiH or H₂O shields more than for non-hydrogenous shields. Further design and material studies would need to be performed to accurately characterize mass differences. One possible reason to promote B to the primary materials list is if it is identified as a lower cost and less hazardous alternate to Be. That study was not performed due to project restructuring.

Table 1: Summary of Optimized Base-Case Reactor Shield Configurations (without pipes) [Reference (d)]

<table>
<thead>
<tr>
<th>Name</th>
<th>Material layers</th>
<th>Thick. (cm)</th>
<th>Mass (kg)</th>
<th>Name</th>
<th>Material layers</th>
<th>Thick. (cm)</th>
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The jugular issues for B are unknown.

2.3. Eliminated Shield Materials

2.3.1. Zirconium Carbide (ZrC)

Zirconium carbide is a good gamma-attenuating material. Zirconium carbide was eliminated due to the expected need for a full materials qualification program, and no obvious mass benefit relative to W (expected to be similar to ZrB₂ in Reference (e)).

2.3.2. Zirconium Diboride (ZrB₂)

A reactor shield is a new application for zirconium diboride. It can attenuate gamma radiation similar but not as well as ZrC, but unlike ZrC, ZrB₂ can also absorb neutrons and moderate neutrons to some extent. This opens the possibility of ZrB₂ performing two roles in the shield. There is little known about ZrB₂, and it is expected to require a comprehensive material qualification program both in and out of pile if used in a reactor.

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shield. Helium generation in ZrB$_2$ would have made qualification more difficult than for ZrC. Preliminary ORNL shield scoping studies did not show a mass savings for shields incorporating ZrB$_2$ relative to comparable primary material shields (Reference (e)). Therefore, ZrB$_2$ was eliminated due to the expected need for a full materials qualification program and no obvious mass benefit.

2.3.3. Plastics

There are many plastics that can be considered for shielding, however, the following general requirements should be met: high hydrogen density, stability over the life of the mission, and high temperature capability. Plastics, (or organics in general), are typically comprised of carbon and hydrogen and in many cases, halogens, oxygen or nitrogen. None of these elements are effective neutron absorbers. Therefore, plastics, without the addition of a low gamma emitting neutron absorber such as boron or lithium, do not represent a low mass option. If B or Li are added, secondary charged particle radiation will increase damage to the plastics. Plastics layered with a neutron absorber such as B or B$_4$C, would likely be an effective neutron shield. However, plastics/organics radiolytically decompose irreversibly (exacerbated if the boron or lithium are incorporated in the plastic), and in many cases have low temperature limits due to low energy barriers to the loss of H$_2$ (discussed in Section 2.3.5). Both processes lead to the evolution of hydrogen gas or methane/ethane, etc. The evolved gasses will pressurize any non-vented canning, likely requiring a pressure relief system with fail-safe features. Further, the loss of hydrogen degrades the plastics ability to moderate neutrons, and could compromise the shield effectiveness, or require additional mass to compensate for the hydrogen loss over time.

Plastics (and organics in general) were eliminated due to irreversible radiolytic damage, the need to accommodate potentially significant off-gassing during the shield lifetime, and the difficulty of keeping plastics cooled. However, research completed on polyisobutylene and oils are discussed below.

2.3.4. Polyisobutylene Rubber (PIB)

The issue of thermal decomposition leads to restrictive temperature limitations in most organics with higher hydrogen density. This issue drove the search for higher temperature capable organics. First, benzene ring based organics, primarily in the form of phenyl-based oils, were studied (discussed further in Section 2.3.5). They were quickly eliminated due to their hydrogen density being much lower than the primary materials (H$_2$O, LiH). Then, NRPCT chemists (Roberson, Vollmer) identified that -$\text{CH}_2$- bonding was likely the cause of the common low temperature thermal decomposition, and that -$\text{CH}_2\text{C(CH}_3)_2$- bonding was known to have much more stable bonds, and therefore a higher temperature capability. The observation led to identification of polyisobutylene (PIB) as a potential high hydrogen density, high temperature capable shielding material. PIB is compared with other polymer structures on the basis of structure type and hydrogen density in Figure 4 and Table 2.

\[ \text{With adjacent } -\text{CH}_2\text{- groups, a } \text{H from one } \text{C can readily bond with a } \text{H from an adjacent } \text{C— both hydrogen atoms have } 1s^1 \text{ orbitals, and can readily form } H_2 \text{ with an ideal } 1s^2. \text{ In the case of a } -\text{CH}_2\text{- bonded } \text{H with a } 1s^1 \text{ orbital being adjacent to a } -\text{CH}_3 \text{ methyl-bonded hydrogen with a hybrid SP orbital, the combination of the two hydrogens is much less favorable, requiring much more thermal energy, and therefore the material appears to have higher temperature capability.} \]

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Jugular issues for PIB are unknown. There is some indication in the literature that this material turns oily under irradiation, which would be a serious design concern if pressure relief of hydrogen was also required. However, this phenomenon has not been studied in detail for PIB.

Despite PIB having a higher temperature capability than other plastics, it was eliminated from consideration primarily due to its irreversible radionlytic decomposition.

**Figure 4: Polyisobutylene and Other Organic Materials**

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2.3.5. Oils

Multiple commercial and "text book" oils were evaluated in the search to identify a high hydrogen density alternative to plastics, which are believed to be limited in maximum temperature. Several oils were identified as having higher temperature capability, but invariably, increased temperature capability translated into lower (and in most cases, unacceptable) hydrogen density. Typical saturated organic plastics would have >11 % hydrogen, but break down (evolve H₂) at much lower temperatures, e.g., ~400 – 450 K versus the benzene-ring-based oils at ~500 – 550 K. An inverse relationship was noted, and verified as reasonable by text books⁴ and NRPCT chemists. On the one hand, high temperature capable oils exist because of the stability of the benzene ring: bi-phenyl, tri-phenyl⁵, quadra-phenyl (all having alternative names, and commercial product names). However, these compounds have significantly lower hydrogen density (~6 – 7 % by mass) than saturated organics (~11 – 14 %), as shown in Table 2. This is a critical parameter for selection of a neutron-attenuating space reactor shield material, since the lower the hydrogen %, the thicker the shield will need to be to slow down fast neutrons. The higher temperature capability of a phenyl comes at the expense of removing hydrogen atoms, forming the benzene rings, where the prior bonds with hydrogen atoms are shared in the carbon ring. Therefore, this group of materials inherently has a lower hydrogen density than a saturated organic, e.g., polyethylene.

Table 2: Hydrogen Density in Oils (bi, tri-phenyls) and Polyethylene

<table>
<thead>
<tr>
<th>Material</th>
<th>Chemical Formula</th>
<th>Overall Density (gm/cc @298K)</th>
<th>Hydrogen Density</th>
</tr>
</thead>
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<tr>
<td>Polyisobutylene</td>
<td>-CH₃-C(CH₃)₂⁻</td>
<td>~1</td>
<td>14.4</td>
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<td>Polyethylene</td>
<td>-CH₂⁻</td>
<td>0.95</td>
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<td>Water</td>
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<td>11.2</td>
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<td>Bi-phenyl oil</td>
<td>(C₆H₅)₂</td>
<td>1.09</td>
<td>6.54</td>
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</table>

Off-gassing radiolytically evolved H₂ from a liquid is complicated, especially in space, where the location of a "bubble" is not certain. This does not appear to be readily achievable without a complicated auxiliary system. Should oil be inadvertently off-gassed, shielding would be lost, which could compromise the ability of the reactor shield to meet attenuation goals. Therefore, this class of materials was eliminated.

2.3.6. Carboranes

Carboranes are boron containing carbon cages with high hydrogen content. They are known to be stable to high temperatures (~1273 K), and given that they contain hydrogen and boron, are of significant interest to the nuclear materials community. An example of a carborane is Poly(carborane-siloxane-acetylene), which is an organic-inorganic hybrid polymer containing B-Si-C-O bonds. However, barring detailed material

⁴ The formation of the benzene ring lowers the potential energy (stabilizes) of the organic compound significantly beyond that of simply adding a third double bond to the hexane ring. This increased stability increases the temperature required to drive off H₂.

⁵ Tri-phenyls are an extension of the bi-phenyl calculation, e.g., (C₆H₅)₃. In fact, middle phenyl groups have 1 less hydrogen, and similarly, benzene has 1 more hydrogen.

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property studies, the configuration of most interest in a shielding application would likely be C₂H₁₂B₁₀, which currently can be purchased in small quantities from Fisher Scientific International Inc.

The irradiation behavior and material properties of carboranes are unknown. The University of Florida has proposed to test the thermal stability of a range of carboranes, as well as determine key physical properties. Considering the developmental status of carboranes, significant effort would be required to qualify them for a reactor shield application.

There is no industrial structure for the production of carboranes. To date, carboranes have been produced in small (very expensive) research batches. The limited availability and high associated cost are jugular issues for carboranes. As a result, carboranes are considered high risk, high cost materials and were eliminated from further consideration. If future research demonstrates desirable irradiation behavior and material properties, this material should be reevaluated for reactor shield use.

2.3.7. Yttrium Hydride (YH₃)

Yttrium hydride has the ability to moderate neutrons and attenuate gamma rays. Preliminary ORNL shield scoping studies did not show a mass savings for shields incorporating YH₃ relative to comparable primary material shields. YH₃ is expected to require materials development as well as irradiated materials property testing, however, detailed studies were not performed. Given that design scoping studies did not show a mass advantage using YH₃, it was eliminated.

2.3.8. Zirconium Hydride (ZrH₂₋ₓ)

Zirconium hydride has the ability to moderate neutrons and attenuate gamma rays. This material has been previously used as a neutron moderator in various nuclear reactor applications (Reference (m)), which suggests that extensive irradiated materials testing has been performed. ZrH₂₋ₓ is not expected to require significant materials development nor irradiated materials property testing, however, detailed studies were not performed. Preliminary ORNL shield scoping studies did not show a mass savings for shields incorporating ZrH₂₋ₓ relative to comparable primary material shields, and therefore it was eliminated.

2.3.9. Uranium-Bearing Materials (Including depleted uranium)

Although preliminary ORNL studies have indicated that various forms of uranium-bearing materials (e.g., depleted uranium, UH₃, uranium-boron compounds, etc.) may be competitive with the primary materials in shielding applications, as a class, these materials were dismissed due to high expected cost and transmutation concerns relative to the primary materials. In particular, ²³⁶U can absorb neutrons and eventually produce fissionable ²³⁹Pu, which can subsequently fission yielding various secondary radioactive materials. In addition, handling material during testing that contains fissile ²³⁹Pu can be a significant concern. The uranium-boron compounds may be acceptable from a transmutation standpoint (studies not performed), however, cost and the need for irradiated materials testing are expected to remain as issues. Depleted uranium was originally specified for SP-100 because it was a slightly better gamma shield material, but was replaced with W due to handling issues and the need to separately can the uranium. Therefore, all uranium-bearing materials were eliminated.

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2.3.10. Borated Stainless Steel

Borated stainless steel is a dispersion alloy, which might provide structural support as well as gamma and neutron attenuation. This potential set of benefits was not explored in detail due to the judgment that structural materials were less of a concern at the outset of the Prometheus program than other shield materials. ASTM standards (A887-87) define specifications for borated stainless steel plate, sheet and strip for nuclear applications, as well as test methods and practices. The NRPCT believes that only limited irradiated materials testing would be required for this material. However, the concern with borated stainless steel is the known production of He within the material, which causes embrittlement. Further research is required to understand the full extent of this issue, as well as whether the embrittlement is a key concern for use of this material given the actual mission conditions (e.g., structural loads may be limiting only during launch prior to the material becoming brittle). Additionally, joining methods for dispersion alloys are not well understood, which could require development effort. Preliminary ORNL shield scoping studies did not show a mass savings for shields using borated stainless steel rather than regular stainless steel or other gamma shield materials (Reference (e)). Given these concerns and uncertainties, borated stainless steel was not included as a primary material. However, as noted previously, the NRPCT did not investigate structural materials to any significant degree, and therefore, this material should be revisited by future researchers during those studies.

3. REFERENCES

(c) ORNL Letter ORNL/LTR/NR-JIMO/05-03. “Multi-layer Radiation Shield Optimization Studies and Simulations”, dated February, 2005.
(f) NRPTCT Letter MDO-723-0048, “The Evaluation of Lithium Hydride for Use in a Space Nuclear Reactor Shield, Including a Historical Perspective, for NR Information”, dated December 9, 2005.
(g) NRPTCT Letter MDO-723-0046, “Space Reflectors Materials for a Prometheus Application”, to be issued.

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Attachment to Enclosure to MDO-723-0049:

Lithium Hydride Irradiation Testing Data Compilation

Authors:
Barri Gurau
Rose Lewis

Reviewed By:
James Nash

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This attachment provides the details included in Figures 2 and 3 in the enclosure. The NRPCT included this information in the shield materials summary letter due to its potential significance in resolving the lithium hydride swelling issue.

Attachment Key

Result Key

0.0 = turned to dust/powder, can rupture, swelling ~20%
0.2 = turned to dust/powder, no can rupture, swelling ~10%
0.4 = sample black, flakey, possible chunks, measurable swelling <3%
0.6 = negligible powdering or stuck to can
0.8 = broke into chunks on sectioning capsule
0.9 = no powdering/intact, some blackening
1.0 = no change

Material Condition Key

P: pressed
C: cast
O: out-gassed

Color Key

No color: Data reported in table was as found in references. In some cases, values (i.e. fluences) were estimated from the information available in order to represent the data visually.

Green boxes: The fluxes of overlapping data points were slightly shifted (i.e. 1e19 changed to 1.1e19 n/cm²) to individually display each point.

Yellow boxes: No information found to approximate values.

Leaked/Contaminated with LiOH Column Key

y: Leaked, specimen exposed to atmosphere (i.e. air) during irradiation testing
x.x: wt% of LiOH purposely and/or accidentally added to the sample before testing
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<th>fast E&gt;0.1 MeV</th>
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References:

(i) Hughes, D.J., R.G. Sache, "Experimental Nuclear Physics Division and Theoretical Nuclear Physic Division Report for April, May and June 1947" ANL-4010.

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CONCURRENCE/DESIGN CHECK FORM FOR DOCUMENT NO. MDO-723-0049  Date: 1/20/06

DOCUMENT TITLE: Space Shielding Materials for Prometheus Application

REFERENCES: ENCLOSES: Shield Materials Information

1. ADSARS: PERMANENT RECORD: Yes X No  Reposit: MFLIB  Corporate Auth KAPL  NR PROGRAM  

Key Words: Space Reactor, LiH, lithium hydride, irradiated materials, material properties, Prometheus

Need to Know Categor REP  
Available Sites:  
Design File Location  

2. DESIGN CHECK

Type of Check  Signature(s)  Comments: (Including Reference to Check Document If Appropriate)

A. No check considered necessary  
B. Check vs. previous results/issues  
C. Checked calculations made  
D. Checked computer input and/or output  
E. Computer Programs approved/qualified  
F. Performed independent audit  
G. Spot checked significant points  
H. Reviewed methods used  
I. Reviewed results for reasonableness  
J. Comparison with test data  
K. Reviewed vs. drawings  
L. Verified procedures  
M. Technical content reviewed  
N. Management verification of adequate review by others  
O. Performed Lessons Learned Search  
P. Used Measurement Uncertainty Methods  
Q. Other Checks (Describe)  

3. CONCURRENCE REQUIREMENTS:

Indicate signatures required by X:

SSP MANAGER  
NUCLEAR ENGINEERING  
REACTOR TH/Mech DESIGN  
REACTION EQUIPMENT  
SPP MECHANICAL  
SPP ELECTRICAL  
FINANCE  
PROJECT OFFICE  
ENERGY CONVERSION  

Cognizant Manager  

(Must be Subsection or Higher for External Letters)

4. AUTHORIZED CLASSIFIER: Reviewed By:  

CLASSIFICATION:  

5. RELATED SUBJECTS:

UTRS Implication (Y/N) N  Design Basis Info. (Y/N) N  
UTRS Doc. # N/A  
Commitment Made (Y/N) N  Commitment Complete (Y/N) N  
Safety Council Review (Y/N) N  Design Review (Y/N) N  

PRE-DECISIONAL - For planning and discussion purposes only
Distribution:

NR
DI Curtis, 08S, For NR Information
TJ Mueller, 08R, For NR Information
S Bell, 08I/8024
JP Mosquera, 08C/8017
CH Oosterman, 08C/8017
JD Yoxtheimer, 08S/8034
JM Crye, 08R

SNR
D. Clapper, 065
GM Millis, 065
DJ Potts, 065
H. Miller, 065

BETTIS
SD Harkness
JE Hack
R Baranwal
W Ohlinger
V Munne
DC Noe
HA Karnes, ZAP 31Q
RS Amato, ZAP 31Q
JL Bowman, ZAP 36E
CD Eshelman, ZAP 36E

PNR
J Andes
JF Koury

KAPL
JW Prybylowski, Bin 96
PF Baladasaro, Bin 111
MJ Wollman, Bin 111
H Schwartzman, Bin 132
F Pineau, Bin 098
M Collins, Bin 098
W Burdge, Bln 132
E Pheil, Bin 132
PA Dilorenzo, Bin 132
SA Simonson, Bin 92
G Newsome, Bin 92
Y Ballout, Bin 92
W Johnson, Bin 157
G Stasik, Bin 132
L Kolaya, Bin 92
DF McCoy, Bin 111
JK Witter, Bin 111
W Gideon, Bin 111
DF Poeth, Bin 92
R Lewis, Bin 92
J Nash, Bin 92
B Lugert, Bin 92
BC Campbell, Bin 92
G Young, Bin 163
J Vollmer, Bin 102
R Najafabadi, Bin 102
G Dansfield, Bin 154
R Grossman, Bin 154
C Regan, Bin 092
SM FILE
ADSARS

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