

## **A Review of Tungsten Heavy Alloy Utilization in Isotope Transport Containers – 13380**

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### **ABSTRACT**

A common requirement for radioisotope transport containers is that they provide both durable and efficient shielding of penetrating gamma radiation. This is the case for transport of both spent nuclear fuel as well as intentionally created radioisotopes for medical or other uses. Tungsten heavy alloy (WHA) provides a unique engineering property set for such shielding - easily surpassing more commonly used lead alloys in both strength and attenuation. This family of alloys contains typically 90-98 wt.% W in combination with transition metals such as Ni and Fe. WHA is manufactured in near net shape blanks by liquid phase sintering of compacted powder shapes to full metallurgical density parts. This powder metallurgy approach is described in its ability to provide excellent material utilization and affords efficient manufacturing of various shapes required for gamma shields or collimators.

WHAs offer very high density (approaching 19 g/cc) in combination with relatively high thermal conductivity, low thermal expansion, ambient corrosion resistance, and can be provided with mechanical properties comparable to many medium carbon steels. As such, they can be machined to complex, damage resistant geometries using common metal cutting tools and methods. WHA additionally provides a lower toxicity alternative to Pb- or U-based gamma shielding. Given the specialty nature of WHA, specific metallurgical characteristics are reviewed to assist shielding designers who may otherwise encounter difficulties locating important alloy selection and fabrication details. Contained within this materials and applications overview are guidelines for WHA component design, alloy selection, and practical machining, finishing, and assembly considerations. The microstructure of WHA is that of a metal matrix composite. This factor has specific implications in the design of components for stress service as well as their protection in the presence of electrolytes. WHA is also discussed in the broader context of materials compatibility, as it is rarely used in isolated monolithic shapes.

Alloy selection for new applications is often made primarily on the basis of density. An alternative strategy to this selection approach is presented which proposes that mechanical requirements for a given shielding use be the primary selection criterion over density. Standard commercial grades of WHA for radiation shielding are defined by specifications such as AMS-T-21014 and ASTM B777. These specifications define 4 density classes of as-sintered WHA. Compositional options, as well as post-sinter processing of WHA, are discussed for shielding components that must exhibit higher levels of ductility or very low magnetic permeability.

In addition to the mechanical advantages over Pb-based shielding, the higher linear attenuation of energetic photons for various grades of WHA (as calculated by the NIST XCOM routine) are presented for selected photon energies of interest to illustrate the shield volume reduction generally possible through the use of tungsten-based shielding. While W provides inefficient attenuation of neutrons in a mixed radiation environment, its secondary role in shielding gamma radiation produced as a result of neutron capture is also described.

## INTRODUCTION

The continuing growth of the nuclear industry – whether in the area of power generation, medical use of isotopes, deep radiography, food sterilization, weapons, or scientific research – involves the periodic transport of significant quantities of highly radioactive substances in various forms. Transport casks must be designed so as to provide for efficient and safe handling in all phases of use. Design and construction must offer fundamental durability as well as conformance to applicable regulations. Wet loading of casks introduces additional design and material property considerations. An essential functional element of every transport cask design is that of radiation shielding. While in some applications neutron attenuation is important, gamma shielding is the primary consideration for the majority of transport casks.

Modern transport casks, whether large or small, are seldom monolithic, but consist of multiple concentric components to meet the functional requirements of the device. The gamma shield in modern casks can take the form of an inner cylinder, which may have one or both ends open. A separate shielding disk would then be employed to close one or both ends. In this manner, the high attenuation component is placed very close to the radioactive contents to be shielded, providing both maximum utilization of the high photon scattering material as well as minimizing the size of the gamma shielding component required for a given geometry cask. Descriptions of overall cask design and function will not be addressed herein so as to protect company proprietary detail. This paper will instead focus on the gamma attenuation aspect of modern cask design and how tungsten-based materials fulfill these requirements.

## THE GAMMA SHIELDING FUNCTION

The principal mechanisms for the attenuation of energetic photonic radiation are the photoelectric effect, Compton scattering, and pair production. Within the energy range of interest for gamma shielding of nuclear waste and/or medical isotopes, Compton scattering dominates [1]. Attenuation consequently relates to the material properties of element Z number and density of atomic packing, the latter reflected in the gravimetric density (given in g/cc or similar). These two material properties allow initial screening of candidate materials for use in cask gamma shielding. Additional screening factors such as mechanical properties, toxicity, and cost can then be applied for identification of the optimum element(s).

It is insightful to use the periodic table as a density map, as it already provides a familiar visual display of Z number. Figure 1 shows the natural elements with densities of 10 g/cc and above. For convenient reference, the density of a typical steel is ~7.9 g/cc. Even this moderate density value eliminates the majority of elements from further consideration. Within this element set is Pb at 11.35 g/cc – the *de facto* standard material for a wide variety of x-ray shielding for many decades. Much of the Pb shielding in use is "hard lead" – the yield strength being increased by alloying with Sn, Sb, and/or other metals. While providing the beneficial effect of increasing the deformation resistance of this very soft metal, alloying detracts from both density and predominant Z-number in such cases.

When the minimum density selection criterion is increased to 17 g/cc as shown in Figure 2, very few practical shielding element choices remain. The reduced set contains only the very dense metals W, the Pt group metals, Au, and U. From this group, additional screening of shield material choices can be

made. U is sometimes used for shielding and collimation based on its availability and ability to be readily cast into large shapes, but its utilization is limited both by its high chemical reactivity (resulting in higher corrosion and toxicity) and susceptibility to unwanted nuclear reactions in the presence of a neutron flux. These factors typically offset its superior combination of density and Z number for modern gamma shield design. Material cost becomes the obvious elimination factor for Au and the Pt group metals.

H																	He
Li	Be											B	C	N	O	F	Ne
Na	Mg											Al	Si	P	Se	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	La	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra	Ac															
Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu				
Th	Pa	U															

Figure 1. Only 21 of the 92 naturally occurring elements possess a >10 g/cc density, limiting candidate gamma shielding options.

H																	He
Li	Be											B	C	N	O	F	Ne
Na	Mg											Al	Si	P	Se	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	La	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra	Ac															
Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu				
Th	Pa	U															

Figure 2. Only 7 of the 92 naturally occurring elements possess a density of 17 g/cc or greater. Six of the 7 elements in this group are disfavored based on high cost, limited availability, or radioactivity.

Despite the fact W experienced a significant global price increases since the early 2005 time frame that continues to this day, it remains the only practical high density metal for gamma shield fabrication in the majority of cases. The benefits of using tungsten can be summarized as follows:

- High radiopacity – due to very high density and relatively high Z number
- Low toxicity – offering a greener alternative to lead or uranium alloy usage
- Durability – strength and hardness make possible deformation resistant shapes
- Low chemical reactivity – simplifying corrosion considerations
- Availability – volume output is available from worldwide reserves
- High thermal conductivity – efficiently transfers heat from decay processes

This set of characteristics is unique to W, as can be further seen in Table 1 in comparison with other metals used in cask construction. W offers a linear absorption coefficient approaching that of DU, but somewhat lower due to inferior Z number. The performance of W in shielding gamma radiation from <sup>60</sup>Co is clearly shown to be superior to that of pure Pb. W expands only minimally in response to temperature rise, thus offering attractive shape stability. In casks that undergo a significant temperature change during a use cycle, gamma shields of W will expand less than surrounding stainless steel structures over the same temperature range whereas a Pb shield would expand greater. In the latter case, a Pb shield would be expected to undergo permanent distortion due to creep. W conducts heat an order of magnitude more efficiently than other cask parts made from an austenitic stainless steel such as alloy 304 – valuable in spreading the heat from the interior to larger area heat dissipation surfaces for improved thermal management.

Table 1. Property comparison of various cask construction materials listed with increasing density.

Element/Alloy	Density (g/cc)	Z	Therm. Cond. (W/m-K)	CTE (ppm/K)	$\mu$ (cm <sup>-1</sup> ) for 1.25 MeV
304 stainless	8.0	(~26)	16	17	0.428*
Pb	11.35	82	33	29	0.667**
U	19.1	92	27	19	1.217**
W	19.3	74	160	4.4	1.076**

\* Modeled as Fe-19Cr-9Ni using Reference 2; \*\* calculated with data from Reference 3.

The use of pure W does however present several limitations when considered for use in bulk radiation shielding components. These all relate to the necessity of powder metallurgy (P/M) fabrication given the melting point of W is 3410C – far too high for traditional melt/cast operations. P/M fabrication of W involves the pressing of fine powder in oversize tooling, high pressure powder compaction, and the solid state sintering of the compact at temperatures typically ranging from 2200 to 2600C. Incomplete densification occurs, requiring further hot working to form fully dense shapes. Common deformation processes include rolling, swaging (rotary forging), and drawing of sintered shapes starting from relatively short sintered blanks. This results in certain limitations that include:

- Shape and size limitations – manufacturing largely constrained to rod, bar, and sheet
- Attainable density – full densification requires thermomechanical processing which is difficult for anything other than the basic shapes listed above
- Limited machinability – resultant microstructure may have limited ductility
- Energy intensity – sintering at temperatures of 2400C and above expensive

These issues were realized early in the history of the commercial production of bulk W shapes.

## A PRACTICAL FORM OF TUNGSTEN FOR BULK SHIELDING

In the late 1930s, a solution to such limitations was developed by Price et al that involved the blending of pure W powder with small amounts of transition metal powders [3]. The resultant powder blend enabled liquid phase sintering (LPS), thus permitting the sintering operation to be performed at much lower temperature, and to full metallurgical density. The resultant family of materials have come to be known as tungsten heavy alloys (WHAs), so as to differentiate them from the older term "heavy metals" as they do not share the high toxicity of heavy metals such as Pb, Hg, and others. WHAs are not

alloys in the true sense, but rather metal/metal matrix composites. This characteristic has specific implications in the design of components for end uses involving stress and/or corrosion service. The two phase composite microstructure forms during LPS via a solution-precipitation mechanism (Ostwald Ripening). The ability to utilize the formation of a high temperature liquid phase and yet not lose shape control in sintering results from the transition metal additions having: (1) very limited solubility in the principal tungsten phase and (2) the transition metal binder (or matrix) phase having appreciable solubility for tungsten. The resultant property set offered by this composite has contributions from each phase, as presented in Table 2. WHAs are very significant materials for gamma shielding manufacture in that they retain many of the desirable characteristics of pure tungsten, yet in a form that is much easier to manufacture to full density and can be produced in a much wider range of sizes and shapes.

Table 2. Property development in the WHA composite.

Desirable properties derived from the W phase	Desirable properties resulting from the matrix phase
High density for gamma attenuation	Densification at practical industrial temperatures
Low thermal expansion	P/M with LPS offers far more size and shape flexibility
High thermal conductivity	Ductile matrix imparts easier machinability
Ambient corrosion resistance	Production of durable shapes simplified

Since the original development of WHA, considerable progress has occurred in alloy design. Modern WHAs provide a unique engineering property set for the radiation shielding community - easily surpassing that of more commonly used lead alloys in both strength and attenuation. These attributes include:

- Linear attenuation coefficient of  $\sim 1.04 \text{ cm}^{-1}$  at 1.25 MeV for a Class 4 WHA [2]
- P/M provides near net shape economy and material conservation
- Size capability ranging from gram to metric ton components
- Strength level comparable to some medium carbon steels
- Relatively high thermal conductivity
- Readily machinable with common shop tools and techniques
- Fasteners can be fabricated of WHA so as to avoid hot spots
- WHA surface offers relatively low chemical reactivity
- WHA can readily be recycled

WHAs typically contain 90-98 wt.% W in combination with transition metals such as Ni, Fe, Cu, and/or Co. Gravimetric densities approaching 19 g/cc are possible, though commercial density classes of WHA defined by specifications such as MIL-T-21014D, AMS-T-21014, and ASTM B777 list a nominal 18.5 g/cc density for a 97 wt.% tungsten alloy as the highest. These standards define four commercial density classes of WHA. Alternate compositions can easily be designed for specific applications. The Ni/Fe ratio of most commercial WHAs is maintained between 2 and 7 so as to avoid embrittling intermetallic precipitation on cooling from sintering. A low Ni/Fe ratio risks  $\text{Fe}_7\text{W}_6$  ( $\mu$  phase) formation whereas a very Ni-rich binder can form  $\text{Ni}_4\text{W}$  on slow cooling. The latter intermetallic can be eliminated by post-sinter heat treatment whereas the former cannot.

While the original WHA developed was a W-Ni-Cu formulation, such alloys have been largely

supplanted by the mechanically superior W-Ni-Fe alloys. The latter ternary alloy system has become the industry standard in recent decades. While the W-Ni-Co ternary offers very high mechanical properties for ordnance applications, the presence of Co precludes its use in applications having a potential neutron flux environments as well as for higher energy accelerator shielding in which photonuclear reactions could occur resulting in  $^{60}\text{Co}$  formation.

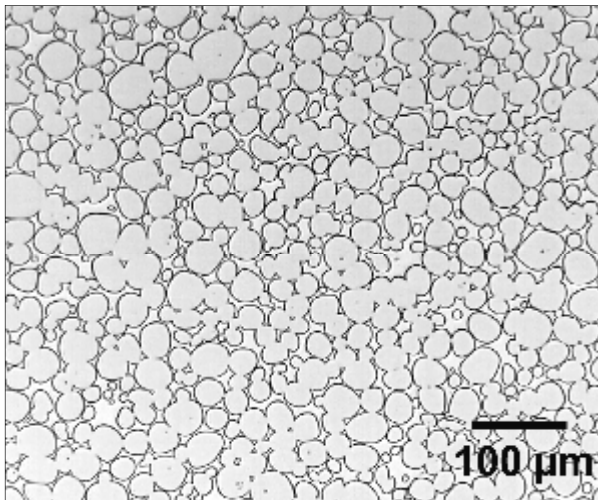


Figure 3. Microstructure of 90W-Ni-Fe (Class 1) WHA showing the W phase spheroidized by LPS. The smooth morphology of this phase is distinctly different than the polyhedral grains commonly seen in pure W microstructure.

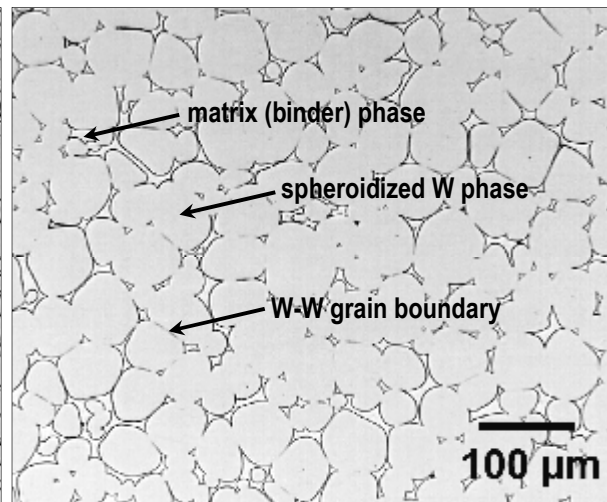


Figure 4. Microstructure of 97W-Ni-Fe exhibits larger spheroid size due to higher LPS temperature and also a higher W-W grain contiguity. The latter factor serves to reduce attainable mechanical properties due to weakness of this interface.

### Current Manufacture of WHA

As is the case for pure W, WHA is likewise manufactured by P/M techniques – again due to the very high melting point of W and now additionally the wide span in melting points of the various alloy constituents. While other shape production options exist, all significant volume production of WHA is by LPS, starting with powder compacted to a specific shapes. This manufacturing approach will now be considered in detail to illustrate some of the considerations that must be addressed up from in the design of large gamma shielding components.

The goal of P/M manufacturing is to capture as much shape detail as possible in oversize press tooling – accounting both for powder compaction and densification during sintering – and preserving that shape through the sintering process to create a full density part requiring little or no additional stock removal. This approach minimizes final part cost by: (1) conservation of high value raw materials and (2) minimizing the amount of stock that must be removed to create the final shape. In the ideal case, the sintered shape will require no additional machining, as net shape provides the best manufacturing economy. However, the reality is that most radiation shielding parts have a geometric complexity and dimensional precision that mandates some secondary machining. The production of near net shape blanks

is common for precision industrial parts requiring very close fit-up.

In that P/M parts are produced in discrete shapes, WHA is not available in mill shapes common to other commercial metals, including long bars, wide area sheet, or coiled product. Maximum blank size is constrained by both pressing and sintering capabilities. When approaching the design of a large shielding array as an assembly of smaller components, it is valuable to consider that P/M favors "blocky" shapes where  $x \sim y \sim z$ . Very long or very thin shapes are more susceptible to breakage during production, and hence increase shield manufacturing cost.

The manufacture of a typical radiation shielding component of WHA starts with very fine ( $\sim 5 \mu\text{m}$ ), high purity powders of W, Ni and Fe. Measured quantities of these raw materials are then mechanically blended to form a homogeneous mix. The blended metal powder is then loaded into press tooling, which may be either of the conventional hard punch-and-die or flexible elastomeric type, the latter being used in cold isostatic compaction (CIP). Isostatic pressing offers the capability to press slabs, cylinders, hollow shapes, and complex curved forms of various sizes – all in the same press cycle. Pressures on the order of 30 ksi are common practice. Considerable powder compaction occurs at this pressure, eliminating much inter-particle air space and forcing interlocking of irregular shaped metal particles to create a pressed part with a sufficient level of handling (or "green") strength. The dynamics of the compaction process and the removal of pressings from tooling are very important factors to be considered early in the design phase of larger radiation shielding components (exceeding  $\sim 500 \text{ kg}$ ). Failure to address these practical considerations will result in shielding components that have an unnecessarily high manufacturing cost due to special handling and yield loss considerations.

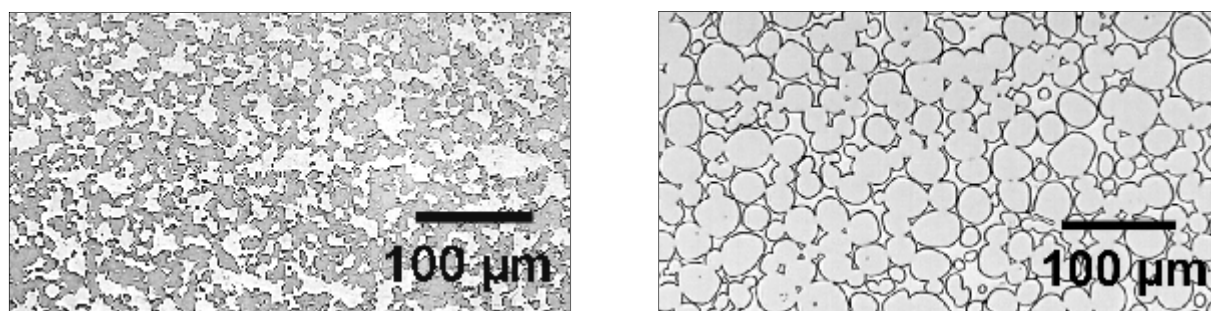
Figure 5. Pressed WHA powder compacts on ceramic media awaiting loading into hydrogen atmosphere stoker furnace. Total door-to-door time for a part on the order of 12 hours.



Powder compacts are subjected to sintering in a  $\text{H}_2$  atmosphere furnace, as shown in Figure 5. While this photo shows a commonly used stoker type continuous furnace, batch type metallurgical furnaces can also be used and are in fact ideal for the sintering of very large radiation shielding parts. As the temperature of a compact increases, the metal oxide layer on each metal particle is reduced by the reactive  $\text{H}_2$  atmosphere, forming water vapor as a reaction product. Optimum heating rate is determined by part section thickness, with massive sections requiring slower heating so as to avoid surface pore closure before the deoxidation process can approach completion. Thermally activated surface diffusion drives inter-particle neck formation in the vicinity of  $1000\text{C}$ . Particle rearrangement and slow

densification occur as this process continues.. As the temperature climbs to ~1400C, the part has achieved near full density.

With time, full density can be achieved in the solid state condition, but the microstructure will not be spheroidized and mechanical properties will be very low even though the gamma attenuation would be no different than from a LPS part. As the part reaches ~1450C, the formation of a liquid metal phase occurs – with the shape of the part being generally maintained by surface tension and an evolving W skeleton. Commercial LPS is performed supersolidus, typically by ~30C, as this promotes a greater fraction of molten binder phase to form – thus aiding spheroidization of the W phase. W particles typically undergo a thousand-fold volumetric growth during the solution-reprecipitation growth sequence. During LPS, a considerable fraction of the W present dissolves in the matrix with 20-25 wt.% W being retained on cooling to room temperature. The Ni-rich matrix phase retains a highly ductile face-centered cubic (FCC) crystal structure. Having a well spheroidized microstructure in radiation shielding components is very important from the perspective of durability. Figure 6 provides both microstructural and tensile property comparisons that emphasize the significance of the LPS structure in WHA.



SOLID STATE SINTERED	CONDITION	MATURE LIQUID PHASE SINTERED
Full	Sintered Density	Full
Poor	Durability / Machinability	Good
<1	Typ. %EL in Tension (As-Sint)	~12
~1	Typ. %EL in Tension (Annealed)	~25

Figure 6. Illustration of the extremes in full density sintering of a Class 1 WHA. While either extreme would provide equivalent gamma attenuation, the SSS condition would offer poor durability whereas the LPS condition would provide high ductility and respond well to post-sinter heat treatment. The sensitivity of tensile elongation to overall quality is evident.

W, like Fe in the ferritic state, possesses a body-centered cubic (BCC) crystal structure. As such, it similarly exhibits a susceptibility for hydrogen embrittlement. In that sintering is performed in a H<sub>2</sub> atmosphere furnace, WHAs supplied in the as-sintered commercial state have mechanical properties limited by the presence of interstitial H. While this is not problematic for many common mass property applications of WHA, it is not a desirable condition for material that will be used in stress applications or for highly mobile radiation shielding. Hydrogen embrittlement of WHA can be essentially eliminated by a post-sinter anneal that permits H outgassing. Material so treated typically exhibits a doubling of tensile elongation, thus promoting "bend before break" type response in critical applications. This may be



especially useful in some very high radiation flux environments where some degree of radiation damage and subsequent loss of ductility is anticipated in metallic bodies. Further mechanical property enhancement is available via a resolution/quench sequence to reduce segregation induced embrittlement that results from trace level interstitial elements. Effective resolution/quench cannot be performed on all sizes and shapes of parts.

Tensile elongation is generally regarded as the most important single measure of microstructural quality of WHA. It indirectly measures both structure and microchemistry. High elongation can only be obtained from high purity, well spheroidized WHA. No amount of post-sinter processing can compensate for structure development that did not mature during LPS. It is also important to note that when sintering in a reactive atmosphere, it is generally not possible to achieve the same level of mechanical properties in large cross section parts as is present in smaller cross section due to differences in both time/temperature history and thermochemistry. This reality likewise implies that in thick section parts, the near-surface region will exhibit higher mechanical properties than material near the center of the cross section. This is an important awareness for scale-up of component designs that were developed subscale.

### **Alloy Selection Criteria**

Materials selection for mass property applications often starts with a specific (sometimes maximum) density as the prime consideration, but that may lead to an unsatisfactory outcome in the case of WHA. A better strategy for many applications is to first define what minimum set of mechanical properties are needed. While some cask designers may start with a given density, the spread in this parameter over the compositional range defined by AMS-T-21014 is relatively small, as seen in Table 3. In stark contrast, ductility can vary a factor of 4 or more in some cases over this limited compositional range – making it a far more sensitive parameter to be considered up front in the cask design process. With only a slight sacrifice in density (and hence linear attenuation coefficient), an appreciable gain in alloy ductility can be realized.

Table 3. Effects of W content for commercially defined WHA compositions.

Variation in wt.% W from Class 1 to 4	7%
Resultant range of nominal alloy density	1.5 g/cc
Resultant difference in tensile properties	typ. $\geq 4X$

While stationary radiation shielding components may require only minimal mechanical properties, shielding arrays for transport casks and other mobile applications may benefit greatly from higher mechanical properties (specifically ductility). This consideration takes on added importance as practical aspects of attachment method, impact resistance, temperature response, and inertial effects of massive parts are considered. These considerations will be significantly influenced by how the gamma shield component is contained within a given cask design, which in turn determines how both stress and heat are transferred within the entire transport cask structure.

The 7 commercial WHA grades defined by AMS-T-21014 are shown in Table 4, with typical measurements being listed rather than specification minima/maxima. Inspection of Table 4 reveals two

groups of alloys: in this case the "standard" Densalloy<sup>®</sup> SD $nnn$  alloys and the "nonmagnetic" Densalloy<sup>®</sup> Dens $nn$  series of alloys. Specifications such as AMS-T-21014 and ASTM B777 define "nonmagnetic" behavior as having a relative magnetic permeability not exceeding 1.05  $\mu$ . As can be noted, the Dens alloys provide a permeability well below this maximum and from a Cu-free composition. While nonmagnetic grades based on the W-Ni-Cu ternary have been used for many decades, the use of a W-Ni-Fe alloy with higher Ni/Fe ratio results in near as low a permeability but providing higher strength and generally better microstructure. Magnetic permeability, while of great significance in shielding used in the proximity of electron optics for high energy x-ray generation, is generally not an important consideration for cask shield design, but may be a useful material option in specialized cases. If a low  $\mu$  alloy is not required, a standard alloy should be selected, as higher as-sintered tensile properties will be realized.

Table 4. Characteristics of ATI Densalloy<sup>®</sup> ternary W-Ni-Fe alloys.

Densalloy <sup>®</sup>	AMS-T-21014 Class	Typical Density (g/cc)/[lbs/in <sup>3</sup> ]	Magnetic Permeability ( $\mu_R$ )*	Use Characteristics
SD170	1	17.09 [.618]	5.0-5.5	Provides greatest ductility and formability
SD175	2	17.62 [.637]	4.5-5.0	Ideal general purpose grade
SD180	3	18.12 [.655]	4.0-4.5	Useful for thick gamma shielding
SD185	4	18.56 [.671]	1.6-2.0	Highest density and gamma attenuation
Dens21	1	17.16 [.620]	~1.01	Greatest ductility in low mu grade
Dens23	2	17.67 [.639]	~1.01	Balance between durability and attenuation
Dens25	3	18.16 [.656]	~1.01	Highest attenuation from low mu grade

\* As measured with Severn gauge ranges, referenced in ASTM A342.

Densalloy<sup>®</sup> is a registered trademark of an ATI group company.

### Mechanical Property Considerations

The industry standard WHA specifications referenced above all call out tensile property requirements based on the testing of coupons, which may be much smaller than actual production parts.. Consequently, the tensile properties of an actual part may vary based on section thickness effects described above, and in almost all cases will be lower. This relationship should be taken into account when developing a material specification and testing plan for a specific WHA shielding component if stress service is an important aspect of its use. The referenced industry standard specifications for WHA call out a minimum set of tensile properties for each density class, as well as reduced property levels required for nonmagnetic grades. As %W is increased, the alloy density and elastic modulus display a systematic increase. Increased %W causes a dramatic drop in attainable ductility and toughness, as the percentage of ductile binder phase is decreased and the statistical number of weaker W-W grain interfaces is increased.

The greatest range of viable processing options exist for lower %W WHAs. These include deformation processing (via rolling, swaging, etc. for shape generation or increased yield strength) and post sinter heat treatment (for reduction of hydrogen and/or segregation induced embrittlement). An illustration of the gain possible in production parts is presented in Table 5, with a 90W-Ni-Fe being used

as the example. While the rather low property set called out by industry specifications for as-sintered coupon properties may be acceptable for many static shielding applications, a significantly higher tensile property set is possible with additional processing. Such properties may be more appropriate for transport containers, where additional stress service considerations may be present. As shown in this table, while strength values are increased only slightly, ductility can be substantially increased, thus reducing the probability of fracture. Hardness of a given WHA changes very little with post-sinter processing for enhanced ductility.

Table 5. Tensile properties of a Class 1 WHA (Densalloy® SD170).

Required ASTM E8, E18 Properties	MIL-T-21014D, AMS-T-21014, ASTM B777	Typical with post-sinter H outgassing of part
UTS (ksi)	110 min.	130
YS, 0.2% offset (ksi)	75 min.	85
EL (1%), 1" gauge	5 min.	28
Hardness (HRC)	32 max.	28

It is again appropriate to comment on the importance of ductility as a factor in materials selection and cask shield design. For casks that are subject to impact loading, it is important that a gamma shield either have high ductility or be protected by a surrounding structure that effectively isolates it from the majority of any impact loading. The W-rich phase of WHA is BCC in crystal structure, and displays strain rate sensitivity. As the rate of stress application is increased, WHA will exhibit higher strength but reduced ductility compared to quasi-static tension measurements. The mechanical dissimilarity between the W phase and the binder phase comprising WHA also gives rise to notch sensitivity. This becomes especially significant in lower ductility and higher %W parts.

### Gamma Shielding Effectiveness

Of primary interest to cask designers is the gamma attenuation behavior of the WHA metal matrix composite. When transitioning a Pb-based shield design to WHA, it must be remembered that for a given attenuation level, the required shield layer thickness will be less while weight will be slightly greater. The combined influences of Z number on mass absorption coefficient and gravimetric density on linear absorption coefficient can be seen in the materials comparison of Table 6. Three photon energies have been selected for comparison: a relatively low 200 keV, the average energy of <sup>60</sup>Co gammas, and a higher energy representative of linac based deep inspection systems. The mass absorption coefficient is seen to undergo a systematic increase with increasing Z number for all energies. Though containing both Ni and Fe, Densalloy® SD180 offers a very similar level of photon attenuation to pure W.

When the linear attenuation coefficient is calculated with density considered, the W-based materials are shown to be clearly superior to Pb-based shielding solutions in reducing gamma shield size. Shielding based on natural or depleted U is superior in both mass and thickness efficiency, but suffers the disadvantages of high reactivity, toxicity, fabrication issues, and susceptibility to unwanted reactions in a neutron environment. In the comparison presented, Densalloy® SD180 provides over 93% (at 1.25 MeV) of the linear attenuation of pure W at slightly reduced density, greater economy, and expanded size/shape

production capability.

Table 6. Comparison of selected gamma shielding materials on both mass and thickness bases (absorption coefficients from NIST [3,5] or calculated using NIST XCOM program [2] with ATI Densalloy® SD180 modeled as 95W-3.57Ni-1.43Fe).

Z	Material	Density (g/cc)	Mass (cm <sup>2</sup> /g)			Linear (cm <sup>-1</sup> )		
			0.2 MeV	1.25 MeV	6.0 MeV	0.2 MeV	1.25 MeV	6.0 MeV
---	SD180	18.12	0.7530	0.0557	0.0416	13.64	1.009	0.753
74	Pure W	19.3	0.7844	0.0558	0.0422	15.14	1.076	0.814
82	Pure Pb	11.35	0.9985	0.0588	0.0439	11.33	0.667	0.498
92	Pure U	19.1	1.298	0.0637	0.0458	24.79	1.217	0.875

WHA provides an efficient gamma shielding material for both fixed and mobile uses that can be machined to precision shapes and has the durability to retain correct shape over time. Maintained shape integrity is especially important for end closures that may suffer deformation or chipping with less durable materials. Table 7 lists TVLs for a range of materials used in bulk shielding applications. These calculations, as are all other linear attenuation calculations in the paper, representative only of narrow beam results and include no other source/shield/detector geometry considerations.

Table 7. TVLs for selected shielding materials (data and calculations from NIST [2,3,5]).

Photon Energy (MeV)	TVL (cm)					
	Ba Concrete	304 Stainless	Pure Pb	SD180	Pure W	DU
1.25 ( <sup>60</sup> Co)	12.7	5.38	3.46	2.28	2.14	1.91
6 MeV	21.7	9.44	4.63	3.05	2.84	2.66

WHA is ineffective for the shielding of neutrons, but may still play an important role in mixed radiation shielding applications. A typical secondary shielding role would be attenuation of 2.2 MeV gamma emissions from H capture of neutrons in PE or similar H-rich layer.

### Thermal Behavior

Radioisotope transport casks are subject to thermal effects both from the external environment as well as internal heating from decay processes. Thermal effects include both changes in material properties as well as thermal expansion. As shown previously in Table 1, W provides a much lower CTE than materials such as austenitic stainless steels that may be used for supportive and/or protective structures. As cask size is increased, the effects of this CTE difference – either in opening gaps or increased thermal stress on components – becomes greater. WHAs average a CTE of ~5 ppm/K – only slightly higher than that of pure W.

Although WHAs contain 90% or greater W, they are not high temperature alloys. While the W phase strongly influences both CTE and thermal conductivity, elevated temperature response is controlled

primarily by the binder phase, which is predominantly a Ni alloy. Figure 7 illustrates the typical tensile property variation that occurs with elevated temperature, as well as subzero. As is typical for most alloys, strength decreases with increasing temperature. In this graph, smooth trend lines were constructed from the discrete data pairs. In reality, if a greater number of tensile tests were performed between ambient and 200F, a discontinuous curve would have resulted for elongation within that interval in that the ductile to brittle transition temperature would have been crossed, which is even more apparent in fracture toughness tests such as Charpy (ASTM E23). This behavior must be considered during cask design to ensure gamma shield durability under frigid transport conditions.

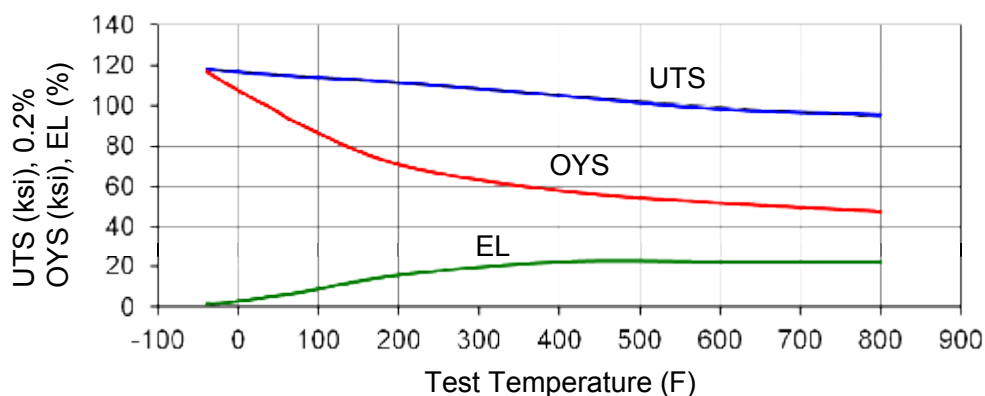


Figure 7. Tensile property variation trends in large cross section Class 3 WHA (Densalloy® SD180) as a function of temperature. All tests performed in open air.

WHAs based on the W-Ni-Fe ternary typically exhibit a thermal conductivity of ~80 W/m-K, easily surpassing that of more common cask materials such as stainless steel and Ni alloys. Decay heat transfer from the interior mass through the gamma shield will accordingly be efficient. Significant temperature differences within a WHA gamma shield would accordingly be minimized.

### Design Guidelines for WHA

As mentioned previously, the nature of P/M fabrication of WHA precludes the option of making very large, single piece shields as would be possible with conventional casting technology. Given the very high density of WHA and the special considerations for large part handling and assembly it presents, this seldom represents a limitation as a large shield would need to be designed as an assembly of smaller, more manageable shapes. As an example, the 18.1 g/cc density of ATI Densalloy® SD180 translates into a practical consideration of safe and effective handling 1130 lb/ft<sup>3</sup> shielding components. Large shields are typically assembled from segments machined to close tolerance for precision fit-up. The use of modular structural shapes additionally provides the option for shield customization without having to modify the entire assembly.

In addition to weight considerations, large transport cask designs exhibit increased sensitivity to differential thermal expansion. The problem for a low expansion WHA gamma shield is reverse that of a Pb alloy shield relative to the CTE for a 300 series stainless steel. Cask designs must maintain gap

closure between structural components so as to maintain adequate thermal transport of decay heat and not allow the opening of gaps that would compromise radiation shielding.

Design, fabrication, and integration of WHA components are not difficult provided the unique characteristics of the material are taken into account. Most significant are the differences of melting point between the component phases and the notch sensitivity resulting from the metal matrix composite microstructure. Good mechanical designs avoid unnecessary stress concentration. This is especially important considering the notch sensitivity of all WHAs. Attention to details such as the sharpness of internal corners, root radii of notches, grooves and threads, and the proximity of holes or other cutouts to part edges help preserve the durability of a WHA part. Concave radii should be limited to 0.020" or greater whenever possible. Holes should not be located closer than 1.5 times the hole diameter from the edge of a part.

## **Shaping**

A principal advantage of WHAs over pure W is their ability to be readily machined into complex geometries using common metal cutting tools and techniques. While it is generally said that WHAs machine similar to gray cast iron, this description can be misleading. Lower %W alloys with high ductility tend to machine more like a stainless steel of comparable hardness. Due to the high elastic stiffness of WHAs, cutting forces will be higher than for most metals. Rigid fixturing and adequate spindle torque are mandatory for good results. The use of a metal cutting coolant/lubricant is optional except where noted. High pH fluids should be avoided.

As the hardness of WHA in most conditions is only 30 HRC or less, it may be readily cut using a heavy duty shop bandsaw equipped with either a bi-metal blade with hook profile teeth or a segmented edge carbide blade at low speed (100-250 sfpm) or alternately by water cooled abrasive cutoff saw. Complex profiles are readily cut in WHA using abrasive (garnet) waterjet. EDM, both wire and sinker type, is routinely used to shape WHA – but as a last resort given the low spark erosion rate of W. Despite this limitation, EDM remains the only practical shaping option for cutting such features as divergent rectilinear windows in shields or collimators. Other common metal cutting techniques such as oxyfuel, plasma jet, and laser cutting should never be used with WHAs. Such methods typically produce unacceptable levels of oxidation and can result in localized thermally induced microcracking.

WHAs are capable of excellent surface finishes when centerless or surface ground. Vitrified bond alumina or silicon carbide wheels of medium hardness are recommended. A water soluble coolant should be used. Diamond wheels should not be used due to rapid loading.

Milling of WHAs is best performed using multi-insert cutter heads. Virtually all commercial WHAs form short chips when machined. The exception to this rule are Class 1 or 2 alloys supplied in a very ductile state, in which case chip breaking must be addressed in tooling selection. Some modern cutter/insert combinations will permit depths of cut on roughing to exceed 0.25" on machines of sufficient power. Best final surface finish is promoted by the use of large nose radius inserts, high spindle speeds, light feed rates, and positive rake inserts. While coated inserts offer improved life when machining most metals, this advantage is sometimes offset when machining WHAs due to the higher cutting forces

created by the rounded (honed) edges necessary for coating of the insert. This should be evaluated on a case by case basis. Recommended milling parameters are provided in Table 8.

Table 8. Suggested milling parameters for WHA.

Milling Operation	Carbide Grade	Rake (°)	Clearance (°)	Edge Condition	Tooth Load (in)	Depth of Cut (in)	Speed (sfpm)
Roughing	K15 or K20	-7° to 0°	0°	honed	0.005-0.015	0.030-0.125	200-400
Finishing	K15 or K20	0° to +7°	0° to -11°	sharp	0.003-0.010	0.005-0.030	300-500

Rotary machining operations such as turning, facing, and boring are likewise straightforward in WHA although special attention must be devoted to the high stiffness of this material. Tungsten carbide cutting inserts are strongly recommended. Attention should be devoted to minimizing chatter, as a WHA workpiece possesses an elastic modulus at least 50% stiffer than a steel toolholder. Optimal insert geometry for both turning and milling will be determined by the specific application. Diamond shapes from 35-80° all function well, with larger angles providing more durable cutting edges. Hexagonal inserts provide further economy for roughing with 6 usable cutting edges, but are restricted from machining narrow features.

Table 9. Suggested turning parameters for WHA.

Milling Operation	Carbide Grade	Rake (°)	Clearance (°)	Edge Condition	Feed Rate (in)	Depth of Cut (in)	Speed (sfpm)
Roughing	K15 or K20	-7° to 0°	0°	honed	0.005-0.020	0.030-0.125	200-350
Finishing	K15 or K20	0° to +7°	0° to -11°	sharp	0.005-0.010	0.005-0.015	250-400

For drilling, standard surface treated HSS twist drill bits generally perform satisfactorily. Very high %W alloys or special high hardness WHAs may mandate the use of carbide bits. As hole size decreases, attention to clearance and the continuous removal of work hardened cutting debris become more critical to avoid seizing or bit breakage. The use of high shear strength tapping lubricant is recommended. A through-spindle coolant provision is useful in larger bit sizes.

Tapping can be the most challenging metal shop operation for WHA due to the high resultant torque on the tap shank. For this reason, 2 or 3 flute, positive rake, spiral point, high clearance taps should be used. Surface treated premium cobalt steel taps typically perform best for this application. The use of a high shear strength tapping lubricant is essential. The coarsest possible thread should be chosen for a given diameter and application. Holes should be tapped to completion without back threading to avoid binding. With care, threads as fine as 2-56 can be successfully tapped. It is generally best to tap large holes with a single point tool. When allowable, the use of a slightly larger pilot hole may simplify difficult tapping situations but will result in slightly reduced thread engagement area. CNC tapping generally provides superior results to manual operations.

## Joining

Joining of WHA either to itself or other metals is an important consideration given that larger

shielding requirements will generally take the form of multi-component assemblies. Whenever feasible, mechanical methods of attachment are preferred, as they are easily inspected, eliminate any possibility of thermochemical alteration of the shielding material, avoid certain material incompatibility issues, and provide known joint strength. Joining is most commonly performed using standard fasteners such as bolts and pins. WHA parts can be likewise be machined with threads or interlocking features to function as fasteners. Standard threaded fasteners can be machined from WHA to replace steel lifting eyes after shield assembly so as to not compromise local radiation attenuation. While ideal for this use, WHA screws or bolt should not be used as lifting or primary attachment fasteners due to the inherent notch sensitivity of all WHAs.

In addition to notch sensitivity, the low CTE (only ~33% of that for 304 stainless) of WHAs must also be considered to avoid fasteners that loosen with repeated (thermal) use cycles. Impact fastening techniques such as impact wrenches and riveting should never be used. Shrink fitting is also a joining option for small assemblies provided the WHA part is the inner member.

As welding of any type involves fusion of the workpiece(s), it is not recommended for WHAs due to the vast difference in both the melting point and CTE between the W phase and binder phase. Microcracking and selective volatilization can occur in the vicinity of a weld.

When a thermal joining technique is required, brazing provides a means of joining WHA to itself or other metals. Brazing is best performed in a hydrogen atmosphere furnace to protect the WHA parts from oxidation and also permit fluxless joining. Compatible filler metals include pure Cu, Monel 400, and standard AWS defined brazing alloys such as BAg-13a. Filler alloys containing S, P, Zn, Cd, or Al should be avoided due to possible interfacial embrittlement. Brazing temperature constraints and the end application generally determine optimum filler metal choice. As with any brazing operation, good joint preparation is essential for producing fully bonded interfaces. Brazing can alter the chemistry within the immediate vicinity of the joint. Points of attachment should not be located along such zones. Manual oxyfuel torch brazing using a flux is also possible but will result in oxidation and is limited to joining very small components. Another useful approach to manual brazing of small assemblies is the use of a TIG (GTAW) torch as a very intense heat source to flow the filler alloy. Leading and trailing inert gas shields promote better protection of the hot WHA against oxidation. Low temperature solders will not wet WHA.

## **General Corrosion Response and Finishing**

WHAs are reasonably resistant to corrosion and are not susceptible to stress corrosion cracking (SCC) as is DU. While ambient corrosion resistance is good, long term corrosion resistance becomes a concern in applications presenting persistent exposure to harsh environments. Such uses would include marine environments and exposure to reactor or holding pool water. On exposure to extreme humidity, salt spray, or the presence of strong electrolytes, corrosion of WHA can occur. This is due to the electrochemical difference between the matrix and the W phases, which sets up micro-scale galvanic cells on the exposed surface. An example is shown in Figure 8. Galvanic contact with other dissimilar metals- especially of greater surface area – provides an additional driver for WHA corrosion. The matrix phase is most readily attacked by acidic solutions whereas the W phase is most rapidly dissolved by alkaline

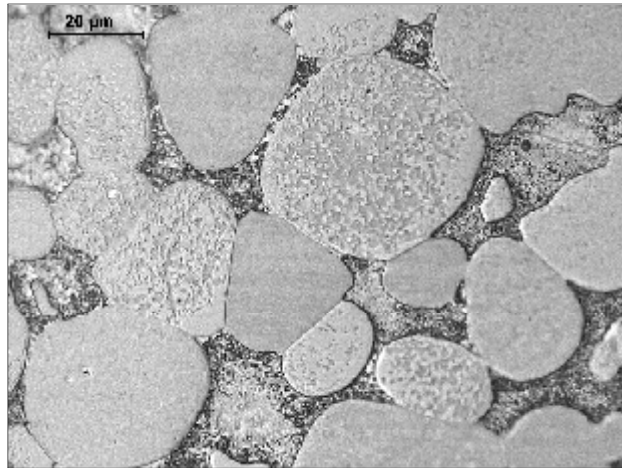


solutions. In cases where such exposure is anticipated, a variety of protective finishes can be applied.

Perhaps one of the most easily applied metallic coatings for corrosion inhibition is electroless Ni. Most electroless Ni platings bond well to WHA, although high P chemistries should be avoided. In corrosion prone applications that also require wear or erosion resistance, a hard Cr plating may offer better protection of the WHA shield element. Both Ni and Cr platings can form a strong bond to WHA.

In many cases, adequate corrosion protection can be provided simply by a strongly adherent polymeric film. A variety of polymeric finishes, including epoxy and acrylic based paints, may be effectively used for corrosion protection. Paints additionally allow convenient color coding and ID marking of components when required, often simplifying cask assembly and QA verification. Organic coatings also provide a dielectric layer, useful in preventing the formation of a galvanic couple. For optimum bond strength, organic coatings should be dried by baking at the recommended temperature to ensure the full set of curing reactions occur.

Figure 8. Example of the aqueous corrosion response of a representative Class 1 WHA (Densalloy® SD170). A polished sample was immersed in a 40 g/L boric acid solution for 90 hours. Corrosion occurred preferentially to the binder phase as expected, with slight orientation sensitive etching of the spheroidized W phase. On the macro scale, this level of corrosion was limited to very slight surface roughening and discoloration. Image shown in reflected light brightfield illumination, original magnification 1000X.



## Conclusions

Tungsten, in spite of its high sustained price in recent years, nevertheless continues to be the element of choice for the manufacture of gamma shielding – whether for large waste transport casks or much smaller point of use shielding for nuclear medicine. Due to both the economy of manufacture and the part size/shape flexibility provided by this fabrication approach, WHAs represent the most practical form of W for use in thick shielding. WHAs offer the shielding designer a unique set of engineering properties that include high radiopacity, a strength level comparable to medium carbon steels, high stiffness, relatively high thermal conductivity, and low surface reactivity under ambient conditions.

This overview has described various aspects of the production of WHA, its metallurgy and resultant properties, industry standard compositions and available property enhancements, and practical consideration for its use in various gamma shielding applications. Awareness of both these material options and limitations is important to the shielding design process for casks, as certain unique properties of WHA such as low CTE become of increasing importance with scale. WHA is readily available in a

variety of forms, and can be customized to adapt to the evolving needs of today's nuclear industry.

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