Review of material issues for the ESS rotating helium-cooled tungsten target

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Abstract. A helium-cooled rotating tungsten target has been retained as the baseline choice for the ESS. This paper presents materials issues associated with the choice of target material and coolant. The operational parameters of the different components of the target are presented to highlight areas where some effort will be directed in future. One of the advantages of going for helium as a coolant compared to water is the possibility to operate without cladding, since no corrosion is expected. However other effects such as erosion of the tungsten surface leading to activated dust circulating in the helium loop, loss of integrity of the tungsten elements or containment of activity could lead to a requirement for cladding. Additional information is required to complement the existing knowledge on several physico-chemical properties of structural and non-structural components of the target. Finally, the basis for the estimation of lifetimes of structural components is reviewed comparing material properties with engineering design requirements.

1. Introduction

A number of target options were reviewed for the European Spallation Source to be built in southern Sweden, with first proton beam on target expected in 2019. The time-averaged 5 MW beam consists of 2.5 GeV protons, delivered in 2.86 ms pulses at a frequency of 14 Hz. The final choice of target material is tungsten, cooled by gaseous helium, with water-cooling being studied as a back-up.

1.1. General parameters

A full review of the Rotating Tungsten Helium-cooled Target RoTHeTa parameters is given in [1]. The target is a 2.5 m diameter rotating wheel with an assembly of segmented tungsten parts enclosed in a structural vessel. The mechanical structure holding the tungsten in place within the wheel will most likely be decoupled from the vessel, the final details will emerge from detailed studies underway.

1.2. Helium coolant parameters

Helium coolant will be circulated at a mass flow rate of 3 $kg.s^{-1}$ and relatively low pressure of 3.5 bar. The helium inlet, outlet and maximum temperatures were estimated to be 20°C, 220°C and 660°C respectively. The flow velocity is 80 $m.s^{-1}$ between tungsten blocks, reaching 400 $m.s^{-1}$ in some areas, which is high and is expected to be lowered considerably through further optimisation of the design.

2. Tungsten as a spallation material at the ESS

Pure tungsten was chosen as the spallation material for ESS rather than alternatives such as tungsten alloys because of operational experience with tungsten at several spallation facilities. This experience is however overwhelmingly with water-cooled assemblies. Online operation showed high corrosion

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rates detected by measuring water resistivity and by sampling the radioisotope content of irradiated water, confirmed by offline measurements of changes in the diameter of irradiated tungsten rods [2]. To mitigate corrosion, various cladding or canning techniques have been developed with materials such as tantalum, stainless steel 304L or alloy 718. Doubts have been raised about maintaining the integrity of tungsten blocks in a helium-cooled spallation target. The issue of tungsten dust production and its eventual contamination of the helium loop is the main topic in this section, which is followed by a discussion of thermo-mechanical behaviour of tungsten and radioisotope release.

2.1. General assumptions for tungsten

The starting assumptions made for tungsten are the following:

- It has no structural role within the target wheel.
- It is brittle.
- No cladding is employed.

Goals for the tungsten parts over the lifetime of the target wheel are expressed as follows:

- They must retain their own structural integrity to keep cooling channels clear for normal and offnormal cooling.
- They must retain as much of the radioactive inventory in the bulk as possible.
- The oxidation rate must be insignificant.
- The manufacturing costs should be kept as low as reasonably possible.

2.2. Tungsten dust production

Assessing the possibility for dust production in a tungsten spallation target cooled with helium is unlikely to yield conclusive trends due to the lack of direct measurements under representative environments: this subject has never previously been considered critical. We offer here a few arguments based partly on general experience with tungsten and partly inferred from data in similar environments. For safety and licensing and for maintainability of the helium loop components, it must be established beyond doubt whether tungsten dust does occur. Since it is not currently possible to answer that question with complete certainty, dust filters are foreseen to be integrated into the helium loop. One important parameter for these filters is the minimum dust particle size, since filtering efficiency of the cyclone filters investigated drops rapidly from 100% to about 85% for submicron particle sizes.

The two possibilities for contamination of the helium loop come from:

a) Pure tungsten, in which case dust size is related to grain size.
b) Tungsten oxide, in which case the size of the volatile oxide particle is unknown.

Concerning a), the grain size is directly related to the heat treatment and forming processes (e.g. rolling) of the tungsten components and the evolution of properties under irradiation. The starting tungsten powder has an average size of 3.85 μm, any grain falling under 0.6 μm is removed from the manufacturing process (for one of the main manufacturers, considered here representative). After sintering and rolling, the grain size grows to between 25 and 200 μm. No evidence exists of pure tungsten dust detaching from the bulk and being entrained in gaseous streams. Perhaps the best example here comes from fusion. It has been argued that the spallation environment would resemble more closely fusion devices than gas-cooled reactors [3]. Yet the fusion community has never considered tungsten dust in helium a problem: whilst the sacrificial layer of tungsten exposed to the harsh plasma environment is studied in great detail for sputtering and blistering effects, no attention is paid to the inner surface of tungsten exposed to flowing helium at 100 bar, 600°C and 200 $m.s^{-1}$ but not exposed to the plasma [4]. The general consensus amongst experts from the fusion community working on tungsten and tungsten manufacturers is that dust from tungsten components is unlikely.

The direct interaction of protons with tungsten is unlikely to lead to the sputtering and blistering effects reported by the fusion community. It has investigated several surface phenomena, establishing thresholds for the onset of blistering and sputtering as a function of incident particle type (hydrogen, deuterium, helium), energy and tungsten temperature [5-7]. The penetration depth of these particles during tests is in the range of a few nm. When converting the high surface fluxes to volumetric particle densities and comparing to the spallation environment (densities of hydrogen and helium), there is a factor 106 difference, i.e. spallation conditions are well below thresholds for the onset of blistering/spluttering.

Concerning b), a few atomic layers of tungsten oxide can be expected to be present on the surface of the as-received tungsten components before operation, but are considered insignificant. Oxidation of tungsten has been observed above 700°C in helium with 5% oxygen. The high operating temperature of the ESS tungsten would lead to significant oxidation if oxygen were present in the environment. However this is very unlikely during normal operation because techniques exist to filter the oxygen from the helium loop to below ppm levels. Furthermore, the addition of 1-5% hydrogen in the helium to reduce any oxides (a practice commonly used industrially with argon atmospheres) could be envisaged, provided this would not interfere with the running of a spallation helium loop, in particular treatment of tritium. If a tungsten oxide layer does eventually grow on the tungsten, it will become volatile from 900°C, leading to a net mass loss as seen in figure 1 where the environment is 25% water vapour in nitrogen. Further studies are required to determine the size distribution of the resulting tungsten oxide particles.

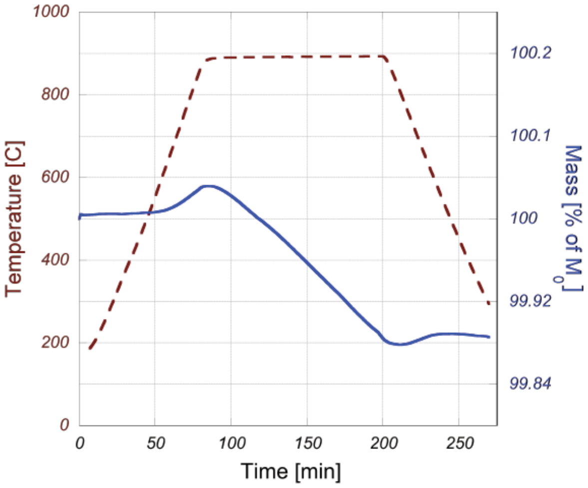

Figure 1: Mass change of a tungsten sample as a function of temperature. The environment is 25% water vapour in nitrogen. An initial increase in mass can be observed due to oxide creation above 600°C followed by net mass loss at 900°C.

A small fraction of the total inventory of irradiated tungsten from spallation environments has been irradiated under a helium environment. At LANL, rod bundles consisting of tungsten rods canned in stainless steel SS304L or Alloy718 tubes using the slip-cladding technique were irradiated, with a 10-20 µm helium-filled gap between each tungsten rod, 2.642 mm diameter, and its enclosure [8]. Rod bundles were irradiated during 6 months, reaching a peak damage of 23.3 dpa. After irradiation, it is reported that the tungsten surfaces showed no indication of any degradation, except for a slight discoloration observed in the rod irradiated to the highest fluence. Peak power density of 2.25 $kW.cm^{-3}$, peak heat flux of 148 $W.cm^{-2}$ and peak fluence of 4×10^{21} $particles.cm^{-2}$ where reached. The peak fluence in particular is higher than that expected at ESS. These irradiated samples are still available, and a careful analysis of the trapped helium in the interstitial space would reveal a host of information on potential dust production and long-lived radioactive species diffusing out of the tungsten and being trapped in the helium gas.

2.3. Tungsten cladding/canning/surface treatment

All studies so far are based on the assumption that the tungsten is bare with direct contact to the flowing helium. Studies of potential surface treatments, canning or cladding will be carried out to identify a solution for ESS. Issues with these are:

- consequences of pulsed thermo-mechanical loads on cladding material and interface to tungsten;
- irradiation effects on mechanical properties/microstructure of cladding materials;
- irradiation effects on bonding between tungsten and cladding. This applies in the case where there is direct bonding and is not necessarily true for slip-cladding or canning where there is a space between the tungsten and the cladding/canning material filled with another material (e.g helium, liquid metal);
- compatibility of cladding material with helium coolant in a spallation environment.

2.4. Thermo-mechanical properties of tungsten

Concerning the evolution of mechanical properties, irradiation data on tungsten are limited; there are indications that material with small deformed grains maintain ductility far better than material with recrystallized grains. It is reported that the yield stress increases by almost a factor 2 after irradiation to 23 dpa for samples irradiated at temperatures up to 300°C [9]. Cracking is observed on the sides of the compression specimens after testing suggesting a decrease in ductility after irradiation. The fusion community studied the consequences of significant transmutation of the initial tungsten to mainly osmium and rhenium [10]. Their concern was that a complete change of crystal structure from the α phase to the σ phase along with shrinkage since the σ phase is more densely packed, and other strains, would produce high tensile stresses. The brittle material could then suffer extensive cracking, and eventually crumbling to powder. These effects were considered significant for an advanced fusion device, a model B facility beyond ITER, where after 5 yrs the initially pure tungsten would become W 75.1, Os 12.8 and Re 11.9. The transition from α to σ is thermally activated with a threshold postulated to be around 1300°C. At ITER, the transmutation problem is not considered an issue since the end of life composition of tungsten parts would be W 98, Re 2 and Os 0.03. At ESS, first estimations show W 98, Re 1 and Os 0.08 for a stationary tungsten target after 5 yrs operation. With much lower transmutation rates and operation temperatures below the thermally activated phase transition threshold, the ESS tungsten is therefore expected to remain in the α phase.

The basic physical-thermal properties such as thermal conductivity, heat capacity and emissivity should be studied as a function of irradiation at different temperatures. Knowledge is also required for the physical-mechanical properties such as thermal dilatation, Young's modulus, Poisson ratio, tensile strength, fatigue and hardness properties. Until detailed information is available, large safety margins on these parameters must be considered in the design of the target.

One important aspect of the operation of tungsten targets is the cyclic thermal load due to the 14 Hz pulsed beam in addition to the rotation of the wheel leading to fatigue. Data are required to demonstrate that failure due to fatigue can be avoided. Each pulse leads to a ΔT of 140°C for the most exposed tungsten component. With a pulse on a given tungsten block every 2 s, and 10^8 cycles over the lifetime of such a block, the endurance limit must be well above the maximum stress amplitude expected due to the cyclic thermal loading, currently estimated to be 50 MPa. The challenge for obtaining useable statistics for fatigue curves from irradiation runs is defining as small as possible a set of samples, since space in irradiation rigs is generally limited.

2.5. Fabrication issues

The manufacturing of tungsten components starts with a selection of powder in terms of grain size distribution. The powder itself is the result of several steps ending with a hydrogen reduction of tungsten oxide WO_{3-x}, which we do not consider significant here beyond controlling impurity levels. Further steps including pressing, sintering and thermo-mechanical treatments will affect the mechanical properties. The industry standard for maximum thickness of rolled plates is 25 mm, which will likely set the upper limit for tungsten component thickness within the wheel. The lower limit will be defined by allowable stresses due to the thermal load on the tungsten parts exposed to the highest power deposition. Properties vary significantly as a function of thickness, which might impose the adoption of one plate thickness throughout the wheel. Annealing could be dropped from the manufacturing process to improve fatigue properties.

2.6. Release properties of tungsten

Release properties of tungsten have not been investigated in detail for the operation regime of the ESS target. They must be understood to estimate contamination rates for the helium loop and to dimension filtering systems. Apart from tritium, the most important contributors to the total activity of the target are all heavy elements close to the atomic number of tungsten including isotopes of tungsten itself, rhenium, tantalum, hafnium and ytterbium. Earlier investigations by the radioactive ion beam community which focuses on efficient extraction of radioactive isotopes from target matrices, i.e. the opposite of what we want to achieve, showed that release of rare earths from tungsten was very poor, when compared to tantalum or hafnium with diffusion coefficients of 5×10^{-12} cm^2.s^{-1}, 10^{-9} cm^2.s^{-1} and 10^{-7} cm^2.s^{-1} respectively at 2000°C [11]. As an example, the release of ytterbium produced in high yield by spallation in tungsten is far below expected from diffusion. This discrepancy is thought to be due to slow surface desorption which must be taken into account when studying release. Indications are that most short-lived heavy radioactive isotopes will decay in the tungsten, whilst longer-lived isotopes will likely remain in the bulk tungsten. The assumption made concerning release of tritium is 100% release. Beyond tritium, it can be expected that a few other isotopes will be released from the tungsten, for example noble gases but this has yet to be determined.

3. Structural material for the target wheel

A stainless steel, SS316L, is the baseline material for the target wheel. Data on the behaviour of this steel in a spallation radiation environment exists. It is also used extensively in nuclear systems for which design rules exist. ESS commissioned a study of the current status and applicability of the RCC-MRx nuclear design code to the ESS spallation environment. The main conclusion was that the code could not be used in its current version for parts directly exposed to the proton beam. It might however be used for other parts, provided a thorough analysis of the radiation spectrum is made and shown to be compatible with the RCC-MRx domain. This domain is currently not well defined in the code, and a modification request has been filed by ESS to address this issue, the expected result being a clear definition of the domain of applicability of the code. Furthermore, the basis for estimating the lifetime of the target wheel must be reviewed. With conservative assumptions for the beam profile, 2D parabolic 140 mm × 50 mm, the maximum damage rate is 2.5 dpa/year leading to a lifetime of 4 yrs using the currently accepted limit of 10 dpa. Because the rotation of the wheel is synchronised with the proton beam impact, the proton beam impinges at the same points on the 33 sectors along the target wheel. Whilst this configuration leads to better cooling of the target, it decreases lifetime compared to uniform horizontal spreading of the proton beam on the whole circumference.

3.1. Zoning the target wheel and shaft

The method of zoning properties for design depending on the radiation damage on the system could be applied to the ESS. Essentially, below a given radiation damage threshold, material properties of unirradiated materials can be used. Beyond a second threshold, design is not possible. Between the two thresholds, material properties from irradiation data must be used. These "borderlines" are very well defined in the RCC-MRx. They are however unknown in a spallation environment.

In order to implement zoning, the following must be defined:

- whether the criteria used to define the limits in the code are applicable in the spallation environment.
- sufficient measurements to define the borderlines.

The first of the above conditions is non-trivial because it implies that the damage mechanisms are the same in a spallation target compared to a nuclear environment. The second condition requires a full irradiation program to map the borderlines for a spallation environment.

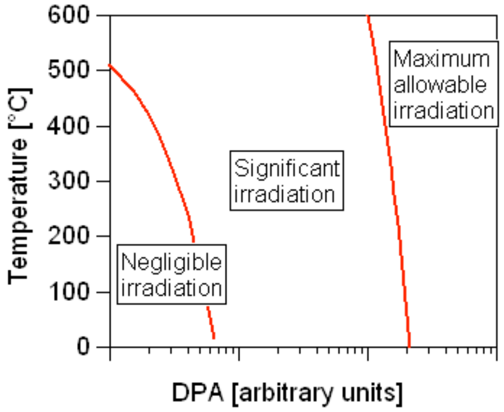

Figure 2: Zoning of irradiation showing where normal design rules can be applied (negligible irradiation), where specific irradiation rules must be applied (significant irradiation), and the maximum allowable limit beyond which operation is not possible.

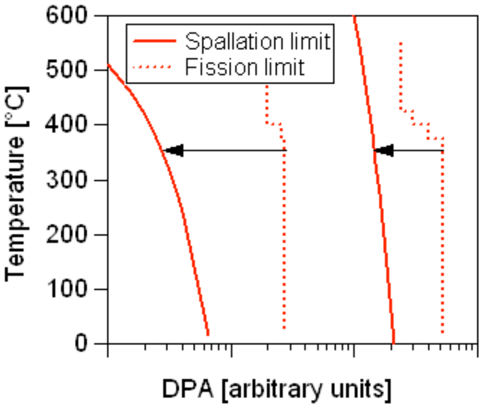

Figure 3: Difference in limits between the fission environment and the spallation environment. Fission limits for design are well characterised, spallation limits are not determined.

Different target wheel areas will be exposed to different particles, fluences and energy spectra. Simulations are underway to provide a precise radiation map of the target station monolith.

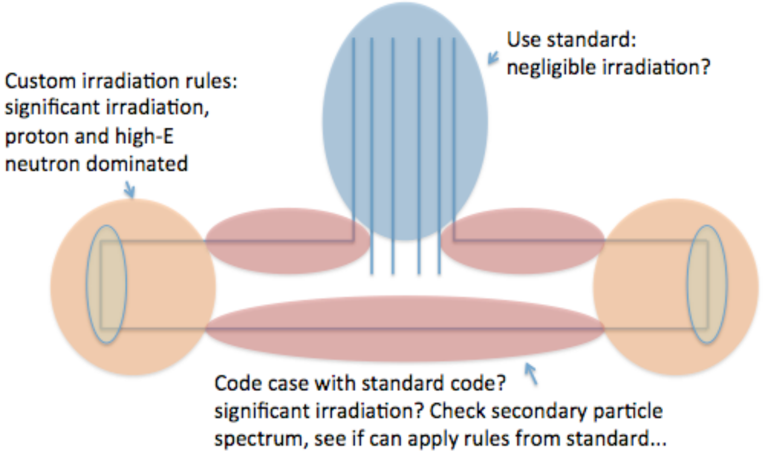

Figure 4: Zoning of the target wheel enclosure and shaft.

3.2. Adding material data to design codes

Several materials are already included in the RCC-MRx code. However some data are missing. It must then be established how additional data can be included in it for operational conditions already covered. For materials not already included in the code, such as titanium alloy Ti6Al4V, data in the negligible irradiation domain must be added. This data largely exists. Data in the non-negligible irradiation domain is scarcer.

For the specific environment of spallation, it is unlikely that any data could be included directly in the code in the near term. However, "code cases" could be drawn, establishing basic rules for design derived from the criteria. Work is needed to understand in more detail the damage modes. As an example, the change in yield strength saturates above a given threshold for the fission environment. It is not clear whether this saturation effect also exists in the spallation environment with a significantly higher helium/dpa ratio.

4. Summary

The development of a rotating helium cooled tungsten target for ESS presents many challenges and opportunities. The challenges lie in the characterisation of structural and non-structural materials under ESS conditions, whilst the opportunities lie in formulating the conditions under which existing design rules can be applied.

5. References

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