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Refractory Metals as Structural Materials for Fusion High Heat Flux Components

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This is a preprint of a paper submitted for publication in Journal of Nuclear Materials
Context within thesis

This paper sets up the argument for using refractories for AM and fusion high heat flux components, highlights new comparative techniques, and includes new data in comparisons.

Novelty

- applying new and previously identified requirements as a set to a fuller range of refractories for this application
- considering suitability by operating temperature range rather than at a single point
- applying a more strengths and weaknesses based approach rather than a simple “in-out” sequence.
- using newly developed tools to present material data and performance metrics
- contributing to the corpus of tools for deciding on the suitability of materials for fusion high heat flux components

Lead author contributions

- consolidation of a number of previous literature reviews and addition of additional references
- extension and formalisation of arguments for revised material selection methodology based on AMAZE project work
- all data selection and presentation
- all writing

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Refractory Metals as Structural Materials for Fusion High Heat Flux Components

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Abstract
Tungsten has long been the favoured armour material for plasma facing high heat flux components proposed for future fusion reactors, but studies examining the use of tungsten or other refractory metals in the underlying cooled structures have historically excluded them, leaving current concepts heavily dependent on copper alloys such as copper chrome zirconium. This paper first outlines the challenge of selecting an appropriate alternative material for this application, with reference to historical selection methodology and design solutions, and then re-examines the use of refractory metals in the light of current design priorities and manufacturing techniques.

The rationale for investigating the suitability of a range of refractory alloys as structural materials is discussed, showing how this is the result of relatively small changes to the logic previously applied, with a greater emphasis on high temperature operation for improved thermal efficiency, a re-evaluation of current costs, a relaxation of absolute activation limits, and the availability of advanced manufacturing techniques such as additive manufacturing. A set of comprehensive qualitative and quantitative assessment criteria are then proposed, drawing on the requirements detailed in the first section; including thermal and mechanical performance, radiation damage tolerance, manufacturability, and cost and availability. Considering these criteria in parallel rather than sequence gives a less binary approach to material selection and instead provides a strengths and weaknesses based summary from which more nuanced conclusions can be drawn.

Data on relevant material properties for a range of candidate materials, including elemental refractory metals and a selection of related candidate alloys are gathered from a range of sources and collated using a newly developed set of tools written in the python language. These tools are then used to apply the aforementioned assessment criteria and display the results. The lack of relevant data for a number of potentially promising materials is highlighted, and although a conclusive best material cannot be identified, refractory alloys in general are proposed as worthy of further investigation.

Keywords: fusion, high heat flux, divertor, materials, refractory

1. Introduction

1.1. The divertor problem

Energy from controlled nuclear fusion promises clean, safe, and abundant electricity and significant advances have been made in recent history, particularly in the field of magnetic confinement fusion, employing the tokamak reactor design. Significant technical challenges remain, however, and commercial viability has yet to be proven. One of the most significant challenges faced by designers of fusion power plants, whatever the technology used, will be extracting heat and exhaust gasses efficiently.

For the tokamak concept, a core element of the power exhaust system is the divertor, where magnetic field lines are directly incident on a region of the vessel wall (the divertor target) and which is subject to steady state heat fluxes in the form of radiation and high energy particles in excess of 10 MW m\(^{-2}\) with excursions due to off-normal events producing transient loads an order of magnitude higher. There is currently no divertor target design suitable for use in a demonstration fusion power plant, either due to insufficient heat handling capability, thermal efficiency, or component lifetime [1]. Significant effort is being spent reducing the heat and particle fluxes incident on the target by adapting the plasma geometry, e.g. [2, 3], but engineering requirements still exceed capability. When compared to designs used for ITER and other current machines, peak incident heat flux, surface material erosion, irradiation damage, and required coolant efficiency are all likely to be higher, in some cases considerably [4].

1.2. Divertor target state of the art

Away from exposed liquid or vapour-based proposals [5] [6] which have a number of significant outstanding technical and physics challenges, leading concepts broadly fall into two categories: water cooled pipes in tungsten monoblocks similar to the design used for ITER [7] and helium cooled thimbles or pipes employing jet impingement [8].

Figure 1 shows two example divertor target designs: ITER-like and HEMJ [HElium cooled Modular Jet-impingement]. The former consists of a water cooled...
CuCrZr pipe with twisted tape insert surrounded by tungsten “monoblocks” as armour and steel mounting blocks as mechanical support. The latter uses a tungsten “thimble” on a tungsten and steel cooled structure, helium coolant, and steel transition pieces to steel support substructure enabling several cooled fingers to be integrated into a single cassette.

![Copper Twisted Tape](image)

**Figure 1:** ITER-like and Helium cooled divertor target designs

A third, high pressure cascade jet impingement concept [10] draws on elements of both of these with a focus on thermal efficiency as well as high performance, but has yet to be fully tested in a representative environment or a manufacturing route proven.

### 1.3. Why focus on the structural material?

As shown above, the fundamental elements of these and other divertor target concepts can broadly be divided into the following sub-components: coolant, armour, cooled structure, and mechanical support.

Pure tungsten or possibly an alloy thereof with additions for ductility or self passivation are generally understood to be the sole candidates for the armour material due to its high melting point, high sputtering resistance, vacuum compatibility, and reasonable resistance to irradiation damage [11], though alternatives including other refractory metals have been considered [12].

The supporting substructure is assumed to be steel, in keeping with divertor cassette designs for ITER [7] and the current DEMO baseline for first wall components, although the design is still evolving and alloys based on zirconium, chromium, and vanadium have also been discussed [13].

Leading candidate coolants include water and helium, with supercritical CO₂, liquid metals, and molten salts considered for more advanced concepts [14]. Hydrogen is used for cooling turbine generators due to its higher thermal conductivity and specific heat, but it has historically been discounted for fusion applications because of embrittlement concerns [15].

Between the armour and mechanical support, the cooled structural material must be joined to the armour and compatible with the chosen coolant, while containing coolant pressure and conducting heat away from the armour. The choice of this material has the largest impact on overall component performance, driving operational temperature, heat flux handling capability, and integration with surrounding interfaces.

Armour material choice is therefore relatively fixed and (beyond facilitating the choice of plasma geometry) the supporting substructure does not have a significant impact on performance of the plasma facing components, but the selection of coolant and the material for the cooled structure remain much more open to innovation and as such provide a potentially strategic avenue to improvements in performance from current designs. In addition, advanced manufacturing techniques, including additive manufacturing (AM), may allow both the use of materials formerly difficult to form and the production of optimised geometries which further enhance performance.

### 2. Structural material selection process

The interfaces between the cooled substructure, the surrounding subcomponents, and the operating environment lead to a raft of somewhat conflicting selection criteria.

Recent material selection processes have focused strongly on thermal conductivity and the avoidance of brittle materials, maximising heat handling capability and ease of fabrication [16, 17]. In addition, a conservative approach to manufacturing risk and the costs of qualifying new materials have further restricted the range of options considered, leading to a narrow reliance on copper alloys such as CuCrZr for water cooled designs and tungsten for higher temperature helium cooled concepts. Refractory alloys other than tungsten have historically featured prominently in attempts to design higher power density concepts, but have ultimately been sidelined due to a strict adherence to activation limits or concerns about hydrogen compatibility [18, 19]. In addition, these studies have tended to make decisions based on performance at a single temperature point, rather than evaluating performance at a range of operating conditions.

As detailed in section 6, radiation by the high energy neutrons produced by fusion causes a wide range of
As detailed in section 1.1, improved thermofluid efficiency will be required for commercial fusion power plants over current concepts. Selecting a coolant temperature above that achievable by the baseline concepts introduces an inherent improvement over current water-cooled baseline. This temperature is also selected to highlight the effect of altered design priorities on the core palette of materials.

The process used in this case begins with mechanical strength at high temperature as well as high thermal conductivity, before applying further restrictions based on activation and then availability, cost and mechanical performance under irradiation.

As a first step. “Low” activation materials are considered to be those for which activation levels are below thresholds as defined in section 6. “Availability, cost and irradiation performance” is a more qualitative filter, enabling a pragmatic exclusion of particularly rare or costly elements or those particularly vulnerable to neutron damage.

The significant difference between this and previous methods is to allow the low ductility of chromium, and moderate thermal conductivity of vanadium, while excluding copper and steel on the basis of their lower melting points. Allowing molybdenum despite its less than favourable activation leaves it as a possible additional candidate and is retained for reasons highlighted later in this paper. It is notable at this stage that such a high-temperature focussed approach naturally lends itself to the refractory metal elements and excludes “traditional” materials including CuCrZr and steels which would otherwise remain the baseline beyond ITER.

A reduced initial threshold temperature would significantly widen the scope of the study, and so focussing on refractory metals as a group gives focus to the exercise and...
facilitates obtaining comparable data.

4. Overview of refractory metals

4.1. Tungsten

With tungsten as the primary candidate for plasma facing armour, including cooling directly would be an attractive option, rather than relying on joints which have historically been the location for part failures under testing, as well as reducing the number of joints to qualify. Tungsten’s inherent brittleness has thus far, however, excluded it from consideration as a structural material, particularly as the ductile to brittle temperature is raised above 800°C under neutron irradiation. Attempts to introduce alloying elements such as Tantalum to increase ductility have shown additional surface modification effects under ion irradiation [20] and no suitable alternative has been discovered. High temperature helium-cooled jet impingement concepts such as the HEMJ design do employ tungsten doped with 1 wt.% La2O3 (WL10) as the impingement surface, though this is supported by a steel substructure [21] and this design requires an enormous number of steel to tungsten welds and a further joint between the tungsten alloy and pure tungsten armour.

4.2. Molybdenum

Molybdenum and its various alloys are used in a range of high temperature structural applications, due to their high thermal conductivity, high strength, and low thermal expansion. TZM (0.5% Ti, 0.08% Zirconium, balance Mo) is one of the most commonly used Mo alloys; its additions of titanium and zirconium act to increase strength and raise the recrystallization temperature. However, TZM in particular is not optimised for use under irradiation and the limited, exploratory irradiation experiments to date have raised concerns over embrittlement [22]. Various developmental alternatives have been proposed to mitigate these effects but are far from commercialisation. It is also important to note that traditional Mo alloys would be suitable only in selected high heat flux locations within a fusion power plant; elsewhere the higher neutron fluxes bring its activation above the permanent disposal waste (PDW) radiological dose limit. Mitigation options for this are discussed in section 6.

4.3. Tantalum

Tantalum has been discussed for fusion applications for more than 20 years [23], and is an attractive prospect for high temperature applications due to its high ductility and corrosion resistance when compared to other refractories, as well as having similarly high strength and melting point. A number of tantalum alloys were developed in the 1960’s for space reactor applications, chief among them T-111 (8% W, 2% Hf, < 100 ppm O, < 50 ppm C, < 50 ppm N, < 10 ppm H) [24] which has been frequently proposed for fusion applications, most recently as a replacement for WL10 in the helium-cooled HEMJ design discussed above. A variant, T-222, with 10 wt% W and 2.5 wt.% Hf, has superior mechanical strength, though available data is less comprehensive [25]. A range of tantalum-tungsten alloys is more readily available, with tungsten percentages ranging from 2.5% (Tantaloy 63) to 10% (Tantaloy 60) with increasing hardness and yield strength and decreasing ductility. [26]

The minimum operating temperature under irradiation is estimated to be well over 1000°C, however, providing a formidable cooling and balance of plant design challenge. Hydrogen embrittlement also remains a significant worry below 600°C even before irradiation. Cost and stability of supply in large quantities have historically been cited as grounds for concern, but are not currently as significant.

4.4. Chromium

Chromium alloys were heavily pursued in the 60s and 70s in Australia, US and Russia as a candidate high temperature material via various alloying and thermal treatment studies [27]. Once a potential rival to the eventually dominant nickel superalloys within the aerospace industry, the chromium alloys ultimately fell out of favour and have found only limited applications in the interim. Interestingly, the fusion community undertook some coarse thermo-mechanical testing of two commercially available Plansee chromium alloys in the early 2000s, before abandoning them citing the lack of room temperature ductility as a barrier to application [28]. Given advancements in manufacturing techniques including AM, this may not remain as significant a hurdle. In addition, the alloys in question (Ducropur — a high purity near-elemental product; Ducrolloy — a 5% Fe alloy to tailor thermal expansion for specific electronic applications) were clearly not developed for to fulfil fusion requirements with the historical chromium studies clearly indicating preferential alloying and/or treatment techniques that successfully offered high temperature strengthening or room temperature ductility superior to the two commercial variants considered.

4.5. Vanadium

There has been considerable interest in using vanadium alloys in fusion applications since the 1990s [29, 30]. Vanadium alloys are inherently promising as a candidate structural material for fusion reactors because of their low irradiation-induced activity, favourable mechanical properties and good manufacturability. V-4Cr-4Ti is currently considered to be the reference V alloy for use in fusion reactors [31]. The nominal operating temperature range for this alloy is of order 600°C to 800°C. However, the supply of this reference material is limited with the largest ingots to date produced in the US, Europe and China but each only representing a few 10s kg. Further challenges are presented with respect to its heat handling
and the prospective integrity of a W-V joint given the moderate thermal conductivity and dissimilar thermal expansion coefficient to tungsten.

5. Thermomechanical performance

For many of the materials under consideration only limited thermophysical data is available and so the following section gives a review of several sources, including historical studies cited above (e.g. [28, 30, 31]), material handbooks (e.g. [26]), internal ITER project technical data (e.g. [32, 33]), and data provided by material suppliers (e.g. [34]). The aim is primarily to draw preliminary performance comparisons (particularly at high temperature), demonstrate the proposed methodology, and highlight key gaps in available information rather than provide absolute conclusions.

The influence of heat treatment and thermal history is significant for all of these materials, and values for both annealed and stress relieved conditions are not always available. Where possible, given the preference for high temperature operation, enhanced ductility over strength (in most cases), and the likelihood of extended periods of time at elevated temperature during operation, the recrystalised values are used. This has a significant impact, for example on the yield stress of tungsten, which varies by a factor of 14 between the recrystalised and stress relieved states at room temperature [33].

As stated above, one of the most significant challenges facing designers of fusion high heat flux components is the lack of fusion-neutron irradiated property data for candidate materials at suitable irradiation temperatures, durations, spectra, and fluence levels [11]. Sections 5.1 and 6 do begin to discuss some of the effects of irradiation on structural materials, but in the absence of suitable irradiated data this section will focus on unirradiated comparisons. The hope is that if promising alternative candidate materials emerge from this new selection methodology, irradiation campaigns will follow.

5.1. Operating temperature window

A temperature window can be defined as a guide to describe limits within which the structural material can best be used [35], although geometric considerations and careful assessment of failure mechanisms must also be taken into consideration. At high temperature, the structural material must retain its mechanical strength, usually limited by thermal creep or increases in environmental interactions as outlined in section 7. At lower temperatures, structural properties are usually reduced by radiation effects, particularly radiation embrittlement, which in BCC materials leads to an increase in the ductile to brittle transition temperature (DBTT) and a loss of ductility.

This window should not, however, be considered to provide absolute limits of operation. The lower limit in particular may be extended if guidance for design with brittle materials is followed, an area of active development for fusion reactor confinement boundaries [36]. Similarly, by careful design and analysis, it may be possible to have parts of a structural component operating at much higher temperature where stresses are less significant or where not exposed to damaging environmental conditions.

5.1.1. High temperature strength

To properly assess high temperature strength, ultimate and yield stress and creep properties across the full temperature range of interest are required, including irradiated properties under fusion neutron irradiation.

Young’s modulus is available for a relatively large number of options, however, and gives a general indication of strength. As figure 3 shows, molybdenum and vanadium are closest to tungsten in absolute terms, but only molybdenum and tantalum retain their strength at temperatures over 800°C.

Figures 4 and 5 show yield stress and ultimate tensile strength for the materials under consideration. As justified above, annealed or recrystalised values have been taken where possible, though like-for-like comparative data for all materials is not available, hence tungsten’s surprisingly low relative yield stress. When considering ultimate tensile strength, TZM’s enhanced ductility over tungsten gives it significant advantages above 600°C and T-111 also shows promise across the full temperature range.

5.1.2. Ductile to brittle transition temperature

At the other end of the temperature window, the ductile to brittle transition temperature defines a lower bound for ideal operation. Design rules for brittle materials such as ceramics in fusion structures are being considered [36]. These are preliminary, however, and ductility throughout the operational temperature range is clearly preferable.
The quantification of radiation induced embrittlement poses an additional challenge, with large uncertainties reported in the change in the DBTT of irradiated tungsten and values up to 880°C used as the baseline [37], leading to the conclusion that at least some brittleness must be anticipated if refractory metals are to be used in the structure.

Directly comparable unirradiated data for all the materials under consideration is not available, and irradiated data is sparse. In addition, fracture toughness measurements are heavily dependent on the details of sample geometry and test conditions.

Data for percentage reduction in area is somewhat more readily available and figure 6 gives values for a number of materials.

This graph shows that in the pure and unirradiated state, of the refractory elements only tantalum and vanadium have particularly attractive ductility at temperatures up to 400°C, with molybdenum a close third. Care should be taken, however, as even moderate quantities of oxygen or other impurities and relatively low levels of irradiation damage have been shown to significantly affect these values [22].

5.2. Thermal stress

5.2.1. Thermal conductivity

With steep thermal gradients inevitable in cooled components subject to high heat flux, understanding and quantifying the interaction between thermal and mechanical performance is critical. Thermal conductivity is the primary driver of this thermal gradient but, as shown in figure 7, apart from copper and its alloys, other candidate materials become increasingly comparable at high temperature.

5.2.2. Coefficient of thermal expansion

High thermal gradients induce high thermal stresses, particularly when joints between dissimilar materials are considered. A lower thermal expansion coefficient generally means a lower thermal stress, but as figure 8 shows, differentiating clearly between candidates on this metric alone is insufficient. The challenge of joining copper or steel to tungsten armour is well highlighted however, it is possible to identify chromium and vanadium as an intermediate group, and it is clear that tantalum and molybdenum alloys are much better matched to tungsten.

5.2.3. Thermal stress figure of merit

In order to clarify these interrelated criteria, Zinkle and Ghoneim [35] define a figure of merit for thermal stress $M$ to qualitatively rank candidate materials, dependent on ultimate tensile strength, thermal conductivity, Young’s modulus, coefficient of thermal expansion, and Poisson’s
ratio. This is related to the maximum allowable heat flux \( \phi_{q_{\text{max}}} \):

\[
\phi_{q_{\text{max}}} \propto \frac{M}{\Delta x} = \frac{\sigma_{UTS} k_{th} (1 - \nu)}{\alpha_{th} E \Delta x}
\]

(1)

Where \( \Delta x \) is the wall thickness of a constrained plate. Thus higher values of \( M \) indicate a better ability for a given material to handle high heat fluxes. Figure 9 shows \( M_{\text{[material]}} \) (normalised against \( M_{\text{tungsten}} \) for clarity) plotted against temperature for a 5mm thick plate in a range of high heat flux materials.

This graph makes it clear why CuCrZr has been such a strong candidate for high heat flux components operating at lower temperatures. However, the significant reduction in strength at high temperature (figures 4 and 5) precludes it from use above about 350°C [37].

Notably, TZM emerges as an attractive prospect, surpassing even CuCrZr, and elemental molybdenum is comparable to pure copper. The remaining materials are much more closely grouped, with tantalum and its alloys only slightly better than chromium, vanadium or even steel.

5.2.4. Thermal mismatch stress

Thermal stress within the structural material due to thermal gradient is not the whole story, however. The primary cause of failure in prototype divertor components has been the interface between the structure (in most cases CuCrZr) and the tungsten armour [38]. Pure copper is used in ITER as a ductile interlayer to mitigate this, and concepts employing more complex compliant or graded structures have also been proposed for DEMO [39, 40]. At the same time, alternative materials with lower thermal mismatches are being explored, including chromium [41]. Applying a thin-walled tube approximation, the stress induced in the structural material due to the thermal expansion mismatch can be calculated using equation 2:

\[
\sigma_{mm} = \alpha_2 (T_{1,\text{mean}} - T_{\text{ref}}) - \alpha_1 (T_{2,\text{mean}} - T_{\text{ref}}) \frac{1}{\frac{1 - \nu_2}{T_2 k_2} + \frac{1 - \nu_1}{T_1 k_1}}
\]

\[
T_{1,\text{mean}} = T_{\text{coolant}} + \frac{q t_1}{h} + \frac{q t_1}{2 k_1}
\]

\[
T_{2,\text{mean}} = T_{\text{coolant}} + \frac{q t_1}{h} + \frac{q t_1}{k_1} + \frac{q t_2}{2 k_2}
\]

(2)

where \( \phi_{mm} \) is the mismatch stress, \( T_{\text{ref}} \) is the reference starting temperature of the component, \( T_{\text{coolant}} \) is the bulk coolant temperature, \( q \) is the incident heat flux, \( t_1 \) and \( t_2 \) are the thicknesses of structure and armour respectively, \( T_{1,\text{mean}} \) and \( T_{2,\text{mean}} \) are the mean temperatures in each
material, $\nu_1$ and $\nu_2$ are Poisson’s ratios, $\alpha_1$ $\alpha_2$ are thermal expansion coefficients, and $E_1$ and $E_2$ are Young’s moduli [42].

Figure 10 shows a graph of the magnitude of this stress with applied heat fluxes up to 25 MW m$^{-2}$ calculated for 1 mm of a range of materials paired with 5 mm of tungsten using 150 °C coolant. Values are not plotted once mean temperatures in the structural material are 100 °C above the maximum temperature for which material data has been provided.

Figure 10: Thermal mismatch stress using 150 °C coolant.

Notably, at a certain combination of heat flux, material thicknesses, and thermal conductivities, using a material with a slightly larger thermal expansion coefficient and lower thermal conductivity than tungsten results in a lower stress at the interface than even a tungsten-tungsten joint, due to the large temperature gradient mitigating the thermal expansion mismatch. At heat fluxes below approximately 7 MW m$^{-2}$ tantalum and molybdenum alloys show the best performance. At higher heat fluxes, however, chromium and vanadium become the clear favourites. The “ideal” heat flux for CuCrZr for this combination of material thicknesses, temperatures, and convective heat transfer coefficients is higher than has been plotted, but the mean material temperature at this point is sufficiently high that structural properties are significantly degraded.

Figure 11 highlights the sensitivity of material pairing to component design parameters. For example, increasing the coolant temperature to 500 °C significantly improves the performance of tantalum and molybdenum alloys, but further excludes copper alloys and moves chromium and vanadium outside their operating temperature windows.

The choice of $T_{ref}$ is also a critical factor in the validity of these graphs, as bonding is usually carried out at elevated temperatures. Geometric design and variation in heat transfer coefficient or methods to remove residual stress during and post assembly (e.g. [43]) will further significantly impact these values, and so these calculations should be taken as demonstrating a figure of merit to be used as a tool for assessing coupled sets of design parameters including material choice rather than as engineering design proposals as they stand.

6. Radiation damage tolerance

As well as radiation induced loss of ductility as described in section 5.1, additional radiation damage mechanisms must be considered, namely activation and swelling.

In order to satisfy environmental responsibility and reduce decommissioning costs, the fusion community have set ambitious and stringent requirements on the activation of power plant components as follows:

As given in [44], for remote handling recycling the dose rate limit is:

$$\sum_{i=1}^{118} A_i X < 10 \text{mSv/hr}$$

and

Decay heat < 10 W/m$^3$

For hands-on recycling the dose rate limit is:

$$\sum_{i=1}^{118} A_i X < 10 \mu\text{Sv/hr}$$

and

Decay heat < 1 W/m$^3$

The above criteria are to be achieved within 100 years ex-reactor.

Combining these and other environmental concerns, IAEA guidelines define a clearance index of a material to determine if the material can be disposed of with no
special precautions. If less than 1 then the material can be disposed of or cleared as if it were non-radioactive [45].

Figures 12, 13, and 14 show dose rate, decay power, and clearance index plots against these limits for tungsten, molybdenum, tantalum, chromium, and vanadium calculated for a neutron spectra corresponding to the first wall of a conceptual fusion power plant [46]. Copper is also included for comparison.

These graphs also show that molybdenum is significantly more activated than the other materials considered, chromium and vanadium are notably less activated, and tantalum compares favourably with tungsten and copper at the 100 year cut-off timescales.

This data shows that molybdenum is significantly more activated than the other materials considered, chromium and vanadium are notably less activated, and tantalum compares favourably with tungsten and copper at the 100 year cut-off timescales.

The currently assumed first wall armour and structural materials of tungsten and copper alloys will not meet this target with the baseline first wall neutron spectra. Given the apparent inevitability of a certain quantity of mid-level waste at the decommissioning stage of the first generation of fusion power plants (or at least for an engineering demonstrator such as DEMO), it seems sensible to tentatively consider the possibility of small quantities of more activated materials during this phase of fusion power development, if concepts employing them provide a route to progressing towards commercialisation and overcoming other challenges in parallel.

If even allowing modest quantities of active waste is discounted, Gilbert et al. [47] have demonstrated that isotope tailoring provides one route to using materials which would not otherwise be palatable, and have used molybdenum as an example. Seven isotopes of molybdenum occur naturally in relatively equal abundance, one of which (Mo97) is significantly less activated than the others. Figure 15 shows that although it does not meet the dose rate requirements, it does compare favourably with tungsten after the 100 year limit.

Achieving the enrichment needed to meet this level of activation will almost certainly be prohibitively expensive for bulk elemental molybdenum, but if quantities of isotopically tailored material are small or if Mo97 is used as a minor alloying element to enhance performance in another candidate material, it may prove a worthwhile tool in the designers’ arsenal and should not be immediately discounted.

The high heat flux and shallow grazing angle of incident particle fluxes on plasma facing surfaces require close geometric tolerances to be maintained throughout the lifetime of divertor components. Helium production in neutron irradiated materials causes swelling [48] which can accelerate surface erosion and induce additional stresses.
Figure 15: Isotope tailoring of molybdenum

which in turn reduce component lifetime. Little data is available for materials other than tungsten in a fusion neutron spectrum, but the BCC crystal structure of the other refractory metals suggests that the degree of swelling will be similar.

7. Chemical compatibility with operating environment

Thermomechanical performance of structural materials for fusion must be maintained throughout the component lifetime while exposed to both the demanding rigours of the tokamak environment on the plasma-facing side and the coolant on internal surfaces. A full evaluation of all the mechanisms involved is beyond the scope of this paper, particularly as comparable quantitative data for each pairing of material and reagent is not available. The following sections, therefore, highlight critical issues and present a brief summary of available knowledge.

7.1. Tokamak gasses

The structural materials will largely be shielded from direct contact with incident particle fluxes by the tungsten armour, but will nonetheless still be exposed to the tokamak environment. Primarily, this means that vapour pressure must be low enough to maintain ultra-high-vacuum, but exposure to hydrogen isotopes and helium pose additional challenges. Tritium retention must be as low as possible in order to both minimise overall radioactive inventory in a reactor and to avoid loss of available fuel due to decay. In addition, exposure to hydrogen causes embrittlement, and this has historically the cause of most concern (e.g. [15, 19]). The degree of embrittlement, particularly when combined with radiation effects, depends heavily on partial pressure of hydrogen and operating temperature of the component. In addition, hydrogen can, in some cases, be sufficiently removed by holding the component at an elevated temperature [49].

7.2. Coolant

Proposed high temperature coolants for fusion high heat flux applications fall broadly into four categories: gasses, supercritical fluids, liquid metals, and molten salts. The choice of pairing of structural material and coolant is an integrated decision closely tied to the geometry and is based on multiple interdependent factors. Comparisons of thermofluid performance, based on bulk coolant temperature, heat transfer coefficient, and critical heat flux have been reported elsewhere, e.g. [14, 50]. Wider balance of plant and reactivity considerations are less connected to the choice of structural material and are also well debated, e.g. [51]. Information is less well collated on issues of corrosion, erosion, and embrittlement in the context of specifically high temperature fusion applications and refractory materials, and so the following sections will focus primarily on these for each type of coolant, while highlighting gaps in knowledge, where necessary.

7.2.1. Gasses

The pressure inside cooling channels will be significantly different from that outside, but compatibility with the leading candidate gas coolants, helium and hydrogen, has been covered above. Other than the embrittlement previously discussed, significant concerns regarding the use of these are generally material independent, and are more operational issues such as leak tightness, cost, thermal efficiency, and safety.

7.2.2. Supercritical fluids

Supercritical water has been proposed as a coolant for both fission and fusion, is currently used in state of the art high temperature fossil fuel power plants up to 600°C, and has been proposed for applications up to 700°C [52]. Despite this, the focus of corrosion studies has been on a relatively small number of conventional materials with less information available on refractory alloys. At first inspection, the high corrosion resistance of refractory alloys suggests that this may not be a significant concern, but the release of both oxygen and hydrogen due to radiolysis under neutron exposure raises the possibility of reaction from both.

There is also some experience of using supercritical CO₂ for gas-cooled fission reactors and it has been proposed for the secondary coolant loop of fusion power plants [53, 54], but its use as a primary coolant has not been fully explored.

In summary, both supercritical water and CO₂ promise high heat transfer coefficients at high temperature at the expense of very high pressure operation and unknown corrosion performance.
7.2.3. Liquid metals

Liquid metals, particularly lithium, lead-lithium, and sodium have persistently been considered as coolants for both fission [55] and fusion [5]. For fusion, this has included use as both coolant and and plasma facing material for blanket and divertor applications (e.g. [56, 6, 57, 58]), to the extent that a lead-lithium divertor was included as the baseline of the most advanced of the European power-plant conceptual study concepts [44].

Dual cooled lithium lead blanket (DCLL) concepts exist for both ITER and DEMO [59, 60]. Liquid metals have very high boiling temperature and high heat transfer capability, even at low pressure, and, if lithium or lead-lithium is used, can perform multiple-duty as coolant, neutron multiplier, and tritium breeder. These significant benefits must be balanced with numerous challenges, however. Magnetohydrodynamic (MHD) effects due to flow in the presence of tokamak magnetic fields increase pumping power significantly, even at low velocities and when care is taken with the orientation of flow channels. Where water is used as a secondary coolant, reactivity in the event of leakages is a significant risk.

As with molten salts, corrosion and erosion are of significant concern. Compatibility with tantalum is well established, but corrosion [61] and other interactions such as wetting properties [62] require further characterisation under fusion-relevant conditions.

7.2.4. Molten salts

Corrosion is the driving factor affecting compatibility between structural material and molten salts. Corrosivity depends heavily on impurities present in the salt used, and so quantitative assessments of compatibility are subject to the need for careful chemistry control. Reviews of the literature suggest that historical studies have mainly been limited to a subset of nickel based superalloys [63], but these give indications of the susceptibility of various alloying elements which can be used to infer relative performance of alloys based on these.

For chloride salts chromium is the most readily corroded element among the most common constituents of nickel alloys, but tantalum chloride has an even lower energy of formation suggesting yet greater vulnerability. Molybdenum, vanadium, and tungsten on the other hand are more comparable to iron and nickel, suggesting performance comparable to alloys previously studied, with molybdenum alloys historically listed as the most favourable for fusion applications (e.g. [23]). Trials using FLiBe with vanadium alloys have also identified tritium permeation and retention as a significant challenge which needs careful control measures [64].

Results for chloride salts reported in the above reviews suggest that they may be more corrosive than their fluoride counterparts, though studies on relevant materials are even more limited.

Although internal coatings can be used to allow use of a wider range of materials, if complex cooling geometries are to be employed, both the application of these coatings and the verification of their efficacy become increasingly challenging.

8. Manufacturing process

Purely selecting a structural material for a high heat flux component on its thermomechanical properties and chemical merits is not sufficient, however: the concept must be manufactured. The cooled structure must be formed into the desired geometry, whether tube or more complex shape, and it must be joined to the armour either directly or using an interlayer. Throughout these processes, the mechanical properties must be maintained—typically meaning the maintenance (or generation) of suitable grain structure.

8.1. Forming

The inherent hardness and high melting points of refractory metals makes them very difficult to form and fragile if complex shapes are used. Drawing for tubes or forging requires high ductility and very high temperature tooling. Tungsten, in particular, is difficult to machine, and cutting using electrode discharge machining is time consuming and expensive. Traditional powder metallurgy including pressing and sintering is limited to relatively simple shapes and does not lend itself to pressure-retaining geometries.

More recent work has focused on tungsten, examining the use of multilayered foils, powder injection moulding, and fibre reinforced composites and seeks to solve some of these problems, particularly striving for increased ductility [65].

Additive manufacturing opens the possibility of employing complex geometries not achievable by conventional manufacturing techniques as well as significantly more efficient use of raw material reducing both wastage and remote handling mass [66]. Figure 16 shows as an example the level of complexity that can be achieved in powder bed additive layer manufacturing, though robust consistency of material properties has yet to be established for a full range of refractory materials, with tantalum and vanadium as the most promising so far [67].

8.2. Joining

As stated in section 5.2.4, The joint between tungsten armour and copper alloy substructure has historically been the primary point of failure for high heat flux components, due to the high thermal expansion mismatch. For these dissimilar metal joints, ductility is a desirable characteristic as detailed above, and the inclusion of a ductile pure copper interlayer is the usual mitigation technique. Newer concepts involving compliant structures have also shown the ability to survive large numbers of cycles at elevated heat fluxes [39]. Novel thermal cycling methods for
brazing dissimilar materials [43] have also been proposed for reducing these stresses in operation.

One of the most active areas of investigation is that of functionally grading the joint. For copper and tungsten, this has been attempted using a range of methods, including plasma spray techniques [68], melt infiltration [69], spark plasma sintering [70], and laminated foils [71]. For refractory metals, wire arc additive manufacturing has been used to produce a promising three layer joint between tantalum, molybdenum, and tungsten, though this work is unpublished at the time of writing.

The mutual solubility of the two materials and any intermetallic phases formed between them are critical factors in the ease of joining, as well as proximity of melting point, if direct melt-based joining processes are to be used without e.g. braze filler. Of the materials under consideration in this paper, tantalum in particular has excellent solubility with tungsten and with its high melting point lends itself well to powder based grading processes.

9. Price and availability

Figure 17 shows the historical price of a number of refractory elements, with copper included for comparison. Price has been a argument for excluding tantalum for consideration, and while costs have fluctuated significantly in recent years, it is certainly an ongoing concern. being nearly an order of magnitude more costly than tungsten. Notably, chromium has remained significantly cheaper than even copper, and molybdenum and vanadium have shown a marked downward trend over the last few years making them worthy of monitoring.

Of ongoing concern is the future availability of candidate materials for fusion applications. Figure 18 shows relative stability over the last 50 years for all materials under consideration, with similar levels of production for tungsten, molybdenum, and vanadium. Tantalum’s relative scarcity and chromium’s significance (e.g. as an alloying element in steel) are also clearly shown.

Concepts based around additive manufacturing require the supply of suitable feedstock. Elemental tungsten, tantalum, molybdenum, and vanadium powders for laser powder bed fusion have been sourced in small quantities and trials are ongoing with the use of both blended and pre-alloyed tungsten-tantalum ratios but quality has been highly variable between suppliers and cost remains high.

In addition, as shown in the proceeding sections, alloys of candidate elements are likely to show better performance overall than the elements alone, and supply chain for fusion-relevant (and in some cases novel) refractory alloys would need to be fully established, including a programme of characterisation and testing, before they could be considered for inclusion as structural material candidates.
10. SWOT analyses

Tables 1 to 5 show one approach for assessing the results detailed above. Each element is subjected to a SWOT analysis summarising the strengths, weaknesses, opportunities, and threats associated with choosing alloys based on it. In this case, “strengths” and “weaknesses” are taken to be inherent properties of the material which are advantageous or disadvantageous for their use. On the other hand “opportunities” and “threats” are external; being either possible research avenues which might prove the material to be useful or factors where there is large uncertainty either due to lack of available data or unknown future priorities.

Table 1: Tungsten SWOT analysis

<table>
<thead>
<tr>
<th>Strengths</th>
<th>Weaknesses</th>
</tr>
</thead>
<tbody>
<tr>
<td>• Current baseline material</td>
<td>• Brittleness</td>
</tr>
<tr>
<td>• No joint to armour needed</td>
<td>• Oxidation</td>
</tr>
<tr>
<td></td>
<td>• Significant loss in strength when recrystalised</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Opportunities</th>
<th>Threats</th>
</tr>
</thead>
<tbody>
<tr>
<td>• Advanced manufacturing may allow cooled structures</td>
<td>• Lack of irradiated data</td>
</tr>
<tr>
<td>• Alloy development may increase ductility</td>
<td>• Variability in quality of supplied material</td>
</tr>
</tbody>
</table>

Table 2: Molybdenum SWOT analysis

<table>
<thead>
<tr>
<th>Strengths</th>
<th>Weaknesses</th>
</tr>
</thead>
<tbody>
<tr>
<td>• TZM has excellent high temperature strength</td>
<td>• Activation</td>
</tr>
<tr>
<td>• High thermal conductivity</td>
<td>• Oxidation</td>
</tr>
<tr>
<td>• Low thermal expansion</td>
<td>• Brittleness</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Opportunities</th>
<th>Threats</th>
</tr>
</thead>
<tbody>
<tr>
<td>• Isotope tailoring may permit some use</td>
<td>• Lack of irradiated data</td>
</tr>
<tr>
<td>• Fusion specific alloys may be possible which are more suitable than TZM under irradiation</td>
<td>• Lack of corrosion data</td>
</tr>
</tbody>
</table>

Lack of available relevant irradiated data is a constant threat across all fusion materials and corrosion data is almost as equally sparse. Although the inclusion in each table might seem excessive, continuing to highlight the strategic relevance of these research areas remains an important task. Similarly, the potential for developing new alloys more specifically suited to fusion applications or revisiting historical research on refractory alloys such as T-111 occurs repeatedly in the “opportunities” quadrants, particularly when seeking to employ advanced technologies such as additive manufacturing with different requirements to traditional techniques.
11. Conclusions

Historical material selection processes for fusion high heat flux structural materials have generally taken a linear sequential approach to requirements, excluding elements from the periodic table or known alloys at each stage. This, along with a prevailing conservatism within the community, has led to a very small pallette of options available to component designers. Some recent work on composites of these materials has shown enhanced strength and damage tolerance, but the potential for significant performance enhancement if alternative materials can be found for cooled structures leads to a desire to re-evaluate the initial material selection process.

Recent advances in manufacturing methods, including additive manufacturing; improved analysis and testing towards design with brittle materials; progress in the practicality of designing alloys for specific applications; and a fuller assessment of the feasibility of isotope tailoring or more pragmatic approach to the rigourous application of the “recyclable in 100 years” mantra will inevitably broaden the aforementioned pallette of candidate materials. Component designers must therefore make choices based on a more nuanced parallel assessment of strengths and compromises, recognising that no “ideal” material exists for any application. Such an assessment will inevitably identify gaps in knowledge and will prompt targeted research, including alloy development, material testing, and irradiation campaigns. New materials, new manufacturing techniques, and new geometric design freedom will also need new design rules for determining structural integrity for future fusion devices and although this need is already recognised for DEMO (e.g. [72]), the early identification of candidate materials and processes will allow their timely inclusion in these design guidelines.

This paper demonstrates one such assessment, focussed on high temperature operation of divertor components, looking at thermomechanical performance, radiation damage tolerance, chemical compatibility, manufacturing, and price and availability of a number of refractory metals identified as a promising group following a preliminary downselection exercise. Material properties and derived figures of merit are compared to current baseline materials and general observations are made at each stage.

Brittleness, manufacturability, and concerns about activation have historically been grounds for excluding many of these refractory alloys from consideration when designing divertor target concepts and other high heat flux components for fusion energy. Despite other issues such as availability and lack of data for alloys rather than pure elements rendering a number of these relatively unattractive at present, the value of further investigation into their development and subsequent use has been discussed.

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References


