TUNGSTEN-BASED MATERIALS FOR DIVERTOR APPLICATIONS - A. F. Rowcliffe (Oak Ridge National Laboratory)

OBJECTIVE

Summarize the current efforts of international fusion programs to utilize tungsten as a divertor material both for ITER and for future DEMO systems. Within this framework indicate how the scientific knowledge and expertise developed within fusion and non-fusion materials programs can be brought to bear to improve the performance of W-based materials designed for operation in the divertor environment.

SUMMARY

This paper summarizes the requirements for W-based materials for the ITER and DEMO divertor environments and the limitations of currently available commercial materials. On-going efforts to understand and to improve the behavior of W-based materials within the international fusion community are summarized together with non-fusion efforts to improve ductility and toughness via nano-scale microstructural development. Theoretical and experimental topics which could form the basis of new initiatives within the US fusion community
include four steps:

a.) Critical analyses of the existing data base,
b.) Fundamental studies of flow and fracture in W and W alloys,
c.) Theory and modeling of radiation damage in these materials, and
d.) Applying alloying and nanoscale strategies developed for other metallic systems.

1. INTRODUCTION

The past 10-15 years has seen significant progress in the understanding of the fundamental mechanisms underlying radiation-induced changes in the microstructure and the mechanical behavior of both metallic and non-metallic systems. Data from experimental programs utilizing fission reactor and charged particle facilities, combined with sophisticated multi-scale modeling, have generated a scientific knowledge base embracing a broad range of topics including defect production and migration, radiation –induced segregation, nucleation and growth of bubbles and voids, phase stability, plastic deformation mechanisms, fracture mechanics behavior and phase stability [1-4]. This knowledge base was successfully utilized in assessing the impact of the ITER thermal and neutron environment on materials properties and to validate the selection of a broad range of structural and non-structural materials [2]. Beyond ITER, the development of advanced concepts for power reactors continues to be heavily dependent on this knowledge base on structural materials which continues to evolve through a network of national and collaborative programs in the US,EU, RF and Japan [1].

Although plasma-facing materials have received only limited attention from the US fusion materials program in recent years, it is apparent that many of the phenomena which strongly impact the behavior and lifetime of the structural materials of the first wall and blanket (FWB) are of equal relevance to the performance of the plasma –facing and heat sink materials of the divertor. The operating conditions of the latter however are even more complex due to the extremely high heat flux conditions coupled with surface phenomena related to erosion and blistering, injection of energetic

alphas and the absorption, migration and release of tritium. Although carbon has been the material of choice over the past 20 years of tokamak development, the transition to ITER foresees the utilization of Be as a first wall armor and the deployment of both carbon fiber composites (CFCs) and W in the divertor. Testing of blanket modules in ITER will see the continued application of Be layers on the FW. However the even more arduous operating conditions of subsequent machines leading to DEMO and subsequent power plants may require the full application of W to all plasma –facing surfaces [5,6].

The most attractive attributes of W are a very high melting point, good thermal conductivity, low sputtering yield and low activation properties in terms of long-term waste management. These advantages are unfortunately coupled with very low fracture toughness characterized by brittle transgranular and intergranular failure regimes which severely restrict the useful operating temperature window and also create a range of fabrication difficulties. Even for temperatures above the ductile-to-brittle transition temperature (DBTT), the fracture toughness of pure W falls within the range of the lower shelf fracture toughness for reduced activation ferritic –martensitic steels (RAFMs). In spite of this, the favored pathway to the development of replaceable divertor components for machines beyond ITER involves a combination of inventive design solutions to accommodate the brittle behavior of W coupled with advanced cooling concepts using high pressure helium. The requirements of high power densities and high operating temperatures for thermal efficiency will dictate a set of operating conditions for a DEMO divertor which will severely challenge the performance of the currently available commercial tungsten products and probably result in unacceptably short component lifetimes [7].The intent of this document is to summarize the current efforts of international fusion programs to utilize tungsten as a divertor material both for ITER and for future DEMO systems, and then to indicate how the scientific knowledge and expertise developed within both fusion and non-fusion materials programs can be brought to bear on the challenging issues of improving the performance of W-based materials for divertor applications.

2. THE DIVERTOR ENVIRONMENT

The surface of the divertor and the underlying heat sink structure present a complex materials–engineering challenge which must be met if a fusion-based energy source is to be successfully developed. Impurity control and ash removal requires that the outer plasma layers are directed out of their poloidal symmetry to interact with the surfaces of the divertor, the interaction being mediated by the properties of the scrape-off layer (SOL). Progress in developing sustainable structures for this environment requires a close interaction between advances in understanding and controlling the properties of the SOL, ingenuity in structural design and both evolutionary and revolutionary advances in materials. At the recent ReNeW Workshop[8], the full scope, complexity and significance of this area of fusion science and engineering were fully explored within Theme 3, ”Taming the Plasma-Materials Interface”. The final report of the Workshop presents a set of research activities or thrusts designed to advance the scientific and technological frontiers of the fusion program over the next two decades The issues related to the plasma-materials interface are embodied in thrusts 9 through 12 and present a cogent overview of the multidisciplinary field of Plasma-Materials Interactions and a sobering assessment of the limitations of currently available W-based materials and their potential for fulfilling the overall long-range goal of developing a viable divertor system for DEMO.

Approximately two decades of research and engineering development within the ITER partnership have culminated in viable concepts for the various segments of the ITER divertor, and the thermal performance of these concepts has been successfully validated by the cyclic heat flux testing of prototype modules. The evolution of this effort is summarized in a series of papers spanning the past

decade or so [9-17]. The development of in-vessel components for ITER was based on the philosophy of utilizing existing commercially-available materials with minor compositional adjustments where needed to met radioactivation requirements. An extensive materials property database was assembled under the auspices of the ITER Materials Handbook with contributions from each of the ITER Home Teams and culminated in the publication of the Materials Assessment Report (MAR) [15]. Section 2.2 of the MAR contains the most comprehensive set of information and properties data that is currently available for pure W and a limited number of W alloys, and covers processing, component fabrication, physical and mechanical properties, thermal fatigue, thermal shock resistance, and radiation effects data.

A summary of the design of the various sections of the ITER divertor, high heat flux (HHF) testing of mock-ups and procurement issues was presented recently by Lorenzetto et al. [16]. The reference ITER design (Fig.1), utilizes water-cooled CFC monoblock for the strike-point region of the vertical target, (steady state heat loads ~ 10 MW/m^2, slow transients ~20MW/m^2), the choice of material being primarily dictated by the capacity of CFCs to withstand high levels of thermal shock without any surface layer melting. The monoblock is bonded to a water-cooled Cu-Cr-Zr alloy heat sink via active metal casting. However, since tritium retention and re-distribution is a serious issue with CFCs [17], tungsten is specified for the upper section of the vertical target (~5 MW/m^2 heat load) in the form of 10 mm thick tiles with a cast Cu inter-layer brazed or HIPped on to the Cu-Cr-Zr heat sink. Prototypes of the CFC sections have survived thermal loads up to 23 MW/m^2 for up to 1000 cycles and the tungsten sections have survived up to 1000 cycles at 10 MW/m^2; both of these conditions are well in excess of the ITER machine requirements. Details of the design and testing of prototypes of the CFC and W prototype components have been published recently [18-20]. Synergistic effects related to radiation damage in W from the fluxes of 14 MeV neutrons and 3.7 MeV alphas are not thought to be critical in ITER since the W does not have a structural function and component replacement is planned after 300 cycles.

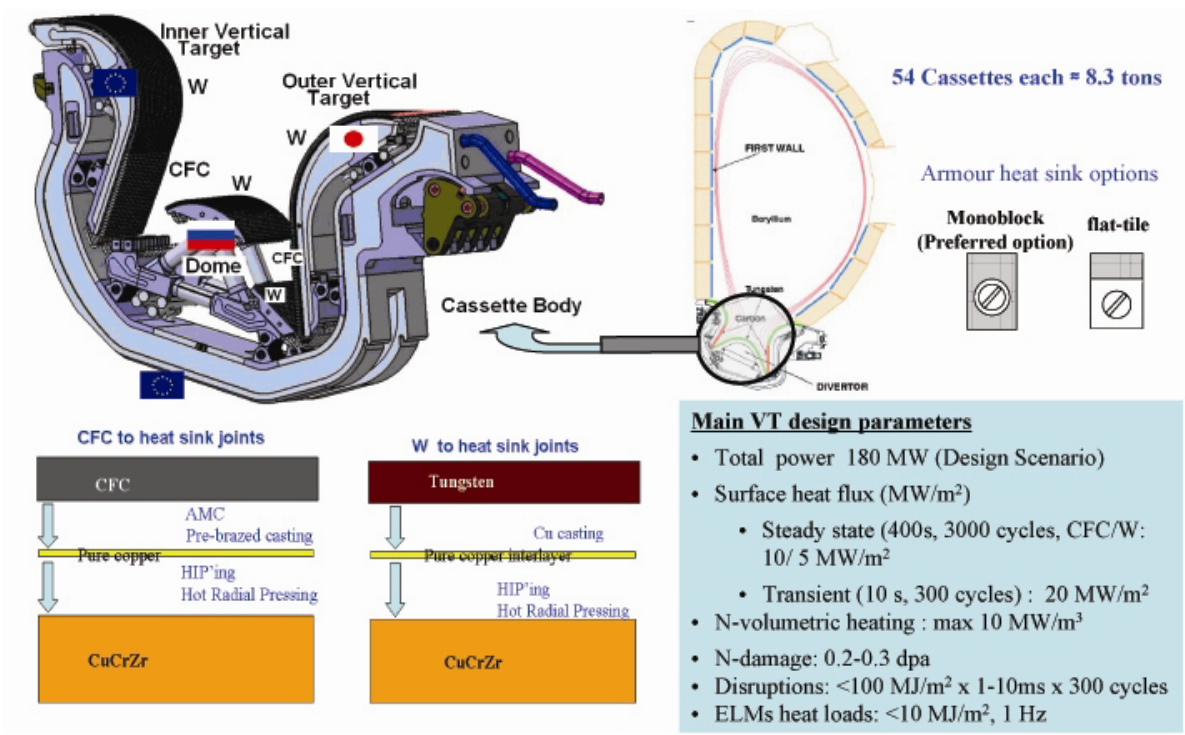

Figure 1. Layout of the ITER divertor and summary of heat loads on the vertical target. From Lorenzetto et al. [16].

Because of the seriousness of the T retention issue in the CFC sections of the ITER divertor, the possibility of converting to an all-W divertor before the start of D-T operations is being debated. Near term and long term research plans on plasma-wall interactions in ITER were presented recently as part of the overall ITER Research Plan (IRP) [17]. Operational experience with a combination of Be, C and W first wall materials are being obtained at JET under the ITER-like wall project [21]. ASDEX-Upgrade has been converted to an all-W wall and recent results indicate reliable operation under ITER-relevant high power/high density discharges with tolerable W concentration levels in the plasma of $< 5 \times 10^{-5}$ and strongly reduced levels of D retention [22].

Since ITER is an experimental machine designed to study the behavior and control of burning plasmas, there is continuing concern regarding the impact of seemingly unavoidable high transient heat loads from ELMs (1-4 MJ/m^2 for 0.3-0.6 ms) and of vertical displacement events (up to 60 MJ/m^2 for 0.5 s) on the integrity of W divertor components. Recent results of surface loadings up to 1.3 MJ/m^2 in the JUDITH-1 e-beam facility have identified various regimes of severe cracking and surface modification as a function of surface temperature and absorbed heat load [23]. The extent of the cracking regime was shown to be dependent on both the specimen orientation in relation to the forging directions and to the final annealing treatment. The implications of off-normal events on the integrity and lifetime of the plasma facing materials have been discussed recently [24,25] and it appears that damage due to surface melting, brittle crack propagation and the erosion of broken particles to form W dust will determine the lifetime of W- armored components in ITER during D-T operation. It is clear that only a very limited number of plasma disruptions could be tolerated without inflicting major damage to the components of the divertor. [25].

Both the steady state operating conditions and possible disruption conditions in machines beyond ITER are even more severe. The operational requirements for a DEMO greatly exceed those of ITER with the plasma materials interface required to remove a four times higher power density and to operate for periods of approximately five times higher duty factor, with surface operating temperatures at ~700°C compared to ~ 200°C in ITER. The continuous reconstitution of the plasma-facing material surface layers by erosion, re-deposition and intermittent melting will necessitate the periodic removal and replacement of divertor components to avoid unacceptable levels of plasma contamination. As a result, goal neutron fluences will probably be considerably lower than for the first wall/blanket but still sufficient (~20 dpa/y) to produce significant property changes to bulk properties, raising the likelihood of synergisms between the bulk and surface related phenomena. For example injection of alphas and tritium at the surface could lead to accelerated degradation of the mechanical integrity and dimensional stability of the bulk material depending on the relative effectiveness of transport and trapping mechanisms for helium and tritium [26-28]. In addition to the overarching issues of thermomechanical loading and surface re-structuring during off-normal events, there is a range of potential bulk and surface damage mechanisms related to the radiation environment which need to be explored and quantified including:

a) radiation hardening-induced shifts in the DBTT,
b) recrystallization during very high temperature excursions and the potential for subsequent brittle intergranular failure regimes,
c) void swelling and the impact of high temperature excursions on subsequent swelling behavior,
d) grain boundary bubble nucleation and growth and the effects of temperature excursions,
e) the fate of injected high energy alpha particles,
f) absorption, transport, trapping and release of T, and
g) nano-scale surface fuzz formation.

Since the DEMO operating conditions are so arduous, it is clear that the experience gained from operating a W divertor in ITER will not provide the full knowledge base needed to allow a transition to a robust design for DEMO. A radically different approach is required, based on high pressure helium cooling coupled with revolutionary improvements in materials performance The huge gaps in our knowledge of the science of plasma-surface interaction and the serious deficiencies in the performance of existing materials which will have to be overcome in order to reach the goal of a power-producing DEMO have been highlighted by Whyte [7]. Some of the critical differences between the divertor parameters of ITER and DEMO are summarized in Table 1. The magnitude of the gaps between near-term and long term divertor concepts are comprehensively addressed, with emphasis on the materials and physics issues, by Raffray et al. [29]. Because of the complexity of the scientific and technological issues involved in the development of a robust divertor system, it has been argued that an intermediate fusion device specifically designed to qualify divertor concepts in a fusion environment will be an essential requirement before moving forward with a DEMO reactor [30, 31].

Table 1. Summary of critical divertor parameters; (from Whyte [7])

Issue / Parameter	Present Tokamaks	ITER	DEMO	Consequences
Quiescent energy exhaust *GJ / day*	~ 10	3,000	60,000	- active cooling - max. tile thickness ~ 10 mm
Transient energy exhaust from plasma instabilities $\Delta T \sim MJ / A_{wall}(m^2) / (1\ ms)^{1/2}$	~ 2	15	60	- require high $T_{melt/ablate}$ - limit? ~ 60 for C and W - surface distortion
Yearly neutron damage in plasma-facing materials *displacements per atom*	~ 0	~ 0.5	20	- evolving material properties thermal conductivity & swelling
Max. gross material removal rate with 1% erosion yield *(mm / operational-year)*	< 1	300	3000	- must redeposit locally - limits lifetime - produces films
Tritium consumption *(g / day)*	< 0.02	20	1000	- Tritium retention in materia and recovery

Design work on a number of helium cooled W divertor concepts has been in progress within the EU and US for several years; details of the designs under development and the related fabrication and joining technologies may be found in references [32-37]. The most advanced design concept from the point of view of progress in the manufacture, qualification and high heat flux testing of individual modules is the European HEMJ design, the main features of which are illustrated in Fig. 2. The design principle is to utilize a small W tile ~5 mm thick as a thermal shield and sacrificial layer; these tiles are brazed to a thimble ~15 mm diameter and ~ 1 mm thick fabricated from W-1%La_2O_3.which forms a cooling finger for helium at 10 MPa and ~ 600/700°C inlet/outlet temperatures. The cooling fingers are

attached to a support structure fabricated from ODS Eurofer steel and the mismatch in thermal expansion between the W and the steel is accommodated by a cast Cu interlock. A summary of the current status of this effort, including design details, materials fabrication, thermal performance analyses and the results of high heat flux testing of individual finger modules, was presented at the IHHF Meeting in 2008 by Norajitra et al. [38].

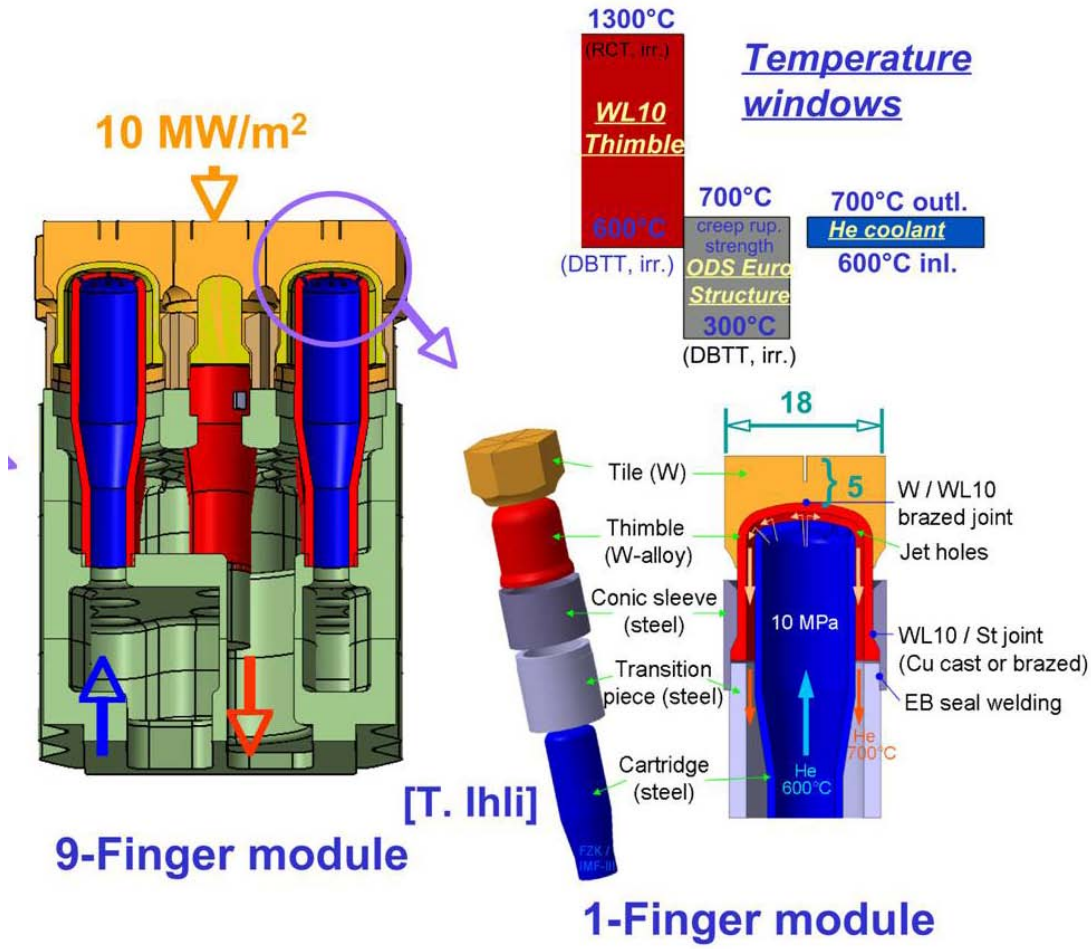

Figure 2. Schematic of the EU helium cooled divertor HEMJ design and the estimated materials operating temperature windows. From Norajitra et al. [35].

Preliminary results of high heat flux testing of prototypes of this and related designs were published recently [39], providing a graphic illustration of the problems associated with using the currently available W materials for this extremely demanding environment. Following high heat flux testing at ~10 MW/m^2 for 70-100 cycles in an electron beam facility, all of the modules suffered serious structural damage (Fig. 3). Subsequent examination revealed:

a) a dense network of cracks perpendicular to the surface extending ~200 microns,
b) coverage of the exposed tile surface with a layer of brazing alloy,
c) cracking of all brazes,
d) propagation of cracks parallel to the elongated grain structure of the W-La_2O_3 and completely through the thimble wall, and
e) development of a gap between the thimble and the sacrificial tile.

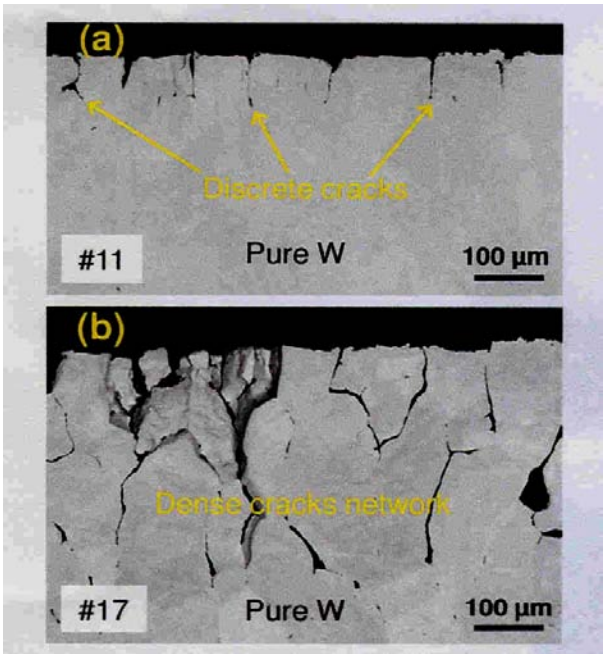

Figure 3. W tile for the HEMJ module, a) surface cracks induced by EDM during manufacture and b) cracking induced during HHF testing for 89 cycles at 10 MW/m^2. From G.Ritz et al. [39]

This experience clearly demonstrates the complexity of the materials issues and design challenges that need to be overcome even without considering the additional deterioration in materials properties induced by neutron irradiation. The application of electro-chemical machining and electro- chemical deposition technologies to mitigate some of the fabrication issues associated with surface cracking and achieving high-integrity brazes was described recently by Krauss et al. [40]

It is generally recognized that to meet the performance requirements of divertor components beyond ITER, innovation in component design and cooling technologies will have to be coupled with dramatic advances in the science and technology of high heat flux materials. Addressing the extreme brittleness of existing commercial rod and plate W materials is probably the most pressing need. Any improvements in the ductility / fracture toughness properties and resistance to recrystallization would have immediate benefits in terms of the ability to survive thermally-induced mechanical stresses without severe brittle failure and also in easing fabrication difficulties. These endeavors must be accompanied by developing an increased understanding of the effects of radiation hardening on flow and fracture in W since any gains in terms of ductility and toughness could be negated by radiation hardening –induced shifts in the DBTT. Any compositional or multi-scale microstructural changes designed to improve fracture resistance will inevitably affect the response to radiation damage.

The additionally important problems related to surface erosion and restructuring during severe thermal and particle fluxes induced by during disruptions, fall primarily into the realm of plasma control and component design rather than the development of new materials. However, the potential for utilizing micro-engineered tungsten, in the form of nano-porous plasma spray or an open cell foam, for armor material in an inertial fusion energy system are being explored [41,42]. More recently, the deployment of W refractory foam materials to enhance heat removal in a flat plate divertor concept has been discussed [43].

3. DEFORMATION AND FRACTURE OF TUNGSTEN

Tungsten in various forms has a wide range of non-nuclear applications, including welding rods, heating elements, lighting filaments, and projectiles. There is a well established fabrication technology based on powder metallurgy and high temperature processing to produce a variety of wire, rod and plate forms. Product consistency in terms of impurity level and grain structure and strength properties is assured via the appropriate industrial standards [44]. The basic physical, mechanical and corrosion properties, details of various processing technologies, alloying behavior (including oxide and carbide dispersion strengthening) are compiled and reviewed in the book by Lassner and Schubert [45]. Theoretical and experimental studies of the fundamental mechanisms of deformation and the influence of microstructural parameters and impurity levels on the DBTT in W and other BCC metals have been extensively pursued for the past 25 years or so and a critical review of this voluminous topic is far beyond the scope of this document. However a thorough appreciation of the materials science of brittle materials, and tungsten in particular, must obviously form the underlying basis of any practical initiatives to moderate the inherent brittleness of W either through alloying or microstructural control. A selection of some of the most relevant contributions to the field of the brittle behavior of BCC materials and of W in particular is provided in references [46-68].

One of the recurring concerns that arise in all high temperature applications of W is the extremely brittle behavior of recrystallized W. There are only limited data on the physical parameters controlling recrystallization, (dislocation density, subgrain structure, texture, time and temperature etc.) and a systematic study has never been attempted. The Group VIA metals in general exhibit intergranular brittle behavior when they are in a recrystallized condition and this phenomenon is generally considered to be related to the behavior of interstitial impurities and their segregation to grain boundaries during the high temperature excursion and subsequent cooling. Molybdenum provides a well known example; Kumar and Eyre [69], considered that the brittle behavior of recrystallized Mo was associated with the segregation of oxygen to grain boundaries and that the effects could be mitigated by the addition of carbon. Earlier Auger fractography studies on W implicated the segregation of P to grain boundaries with the concentration of P increasing with the grain size [70]. More recently, the effects of impurities on the intrinsic fracture resistance of grain boundaries in W have been investigated using a molecular dynamics simulation, Grujicic and Zhao [71] and Grasco [72]. These calculations show that B, C and N strongly increase the cohesive strength of grain boundaries whereas O, Al and Si do not significantly affect the ideal work of decohesion or crack blunting processes. However, both P and S are shown to significantly lower the work of grain boundary decohesion and impede crack tip blunting processes. The fundamentals of grain boundary segregation and the impact of segregation on grain boundary cohesion and fracture were reviewed by Hofmann and Lejcek [73].

4. RADIATION EFFECTS

The data base on radiation effects in W is very limited since given the brittle behavior, there has been very little interest in W-based materials in the nuclear field except as a potential target material for spallation neutron sources [74]. In fact even the basic dose and irradiation temperature dependence of radiation hardening have yet to be determined. Most of the available radiation effects data were collected and summarized in the ITER Materials Assessment Report [15], and subsequently there has been very little added to that database. It is known that neutron irradiation at temperatures below ~500°C to low doses (1.0-1.5 dpa) produces large increases in yield stress and a complete loss of total

elongation. Using data from bend bars and tensile specimens, severe embrittlement has been observed in W and several alloys irradiated to doses below ~1 dpa at temperatures below ~ 400°C (Fig. 4).

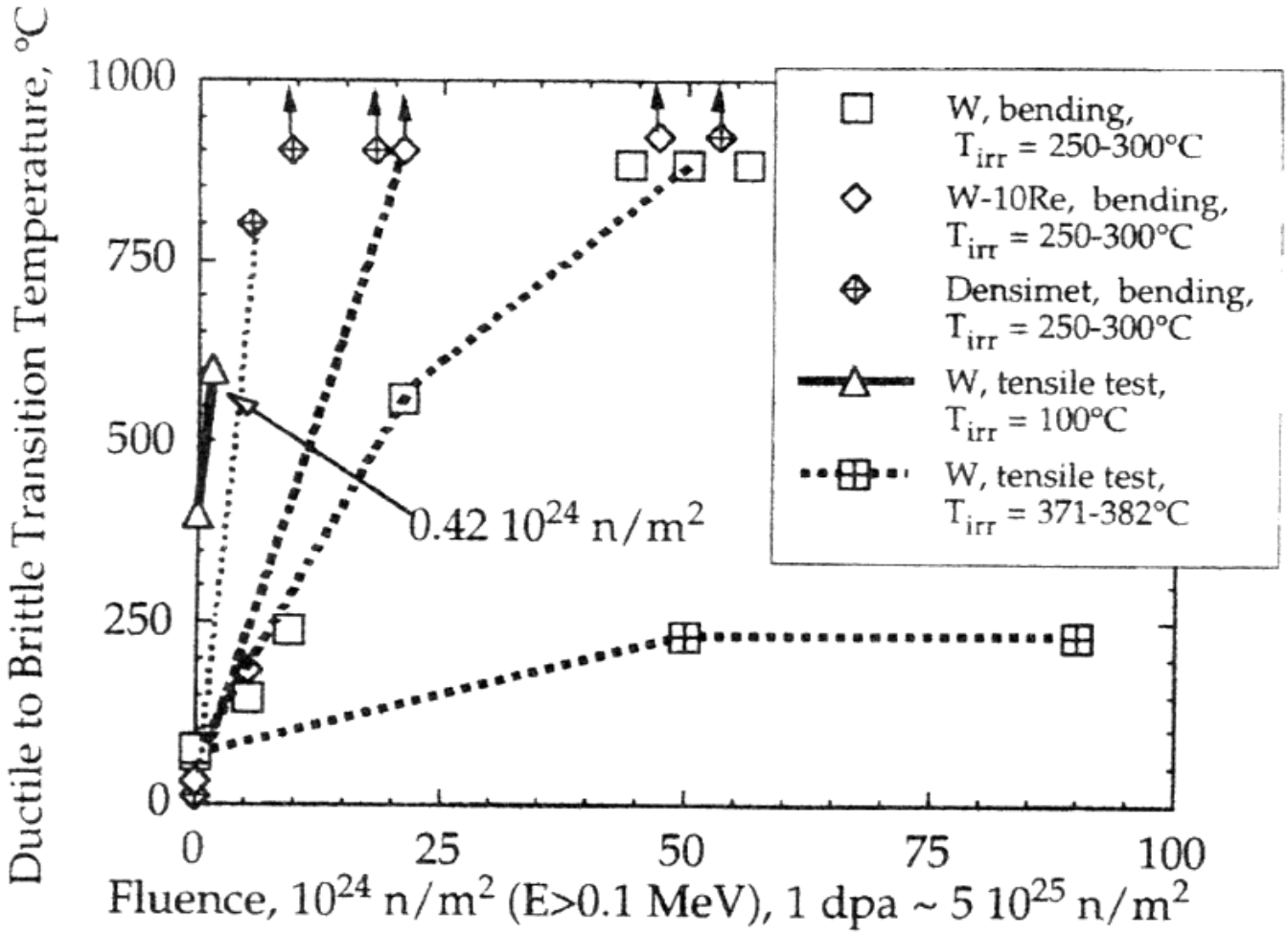

Figure 4. Neutron dose dependence of the DBTT assessed from tensile and bend tests for W, Densimet, and W-10%Re. From the ITER Materials Assessment Report [15].

The minimum irradiation temperature which will avoid serious radiation embrittlement for doses appropriate for DEMO or even for ITER are not known. However, based on available measurements and extrapolating from the data for Mo and TZM, this temperature is probably in the region 800-900°C. The available neutron irradiation data on several W alloys indicate a peak swelling temperature of ~ 800°C for doses up to ~ 9 dpa (Fig. 5) and the formation of void lattices in the range 550-750°C. The behavior of point defects in nanostructured metals during irradiation was discussed by Wurster and Pippan [74] together with a summary of recent experimental data for W-based materials. Additional information on radiation effects may be found in Refs. [75-80]

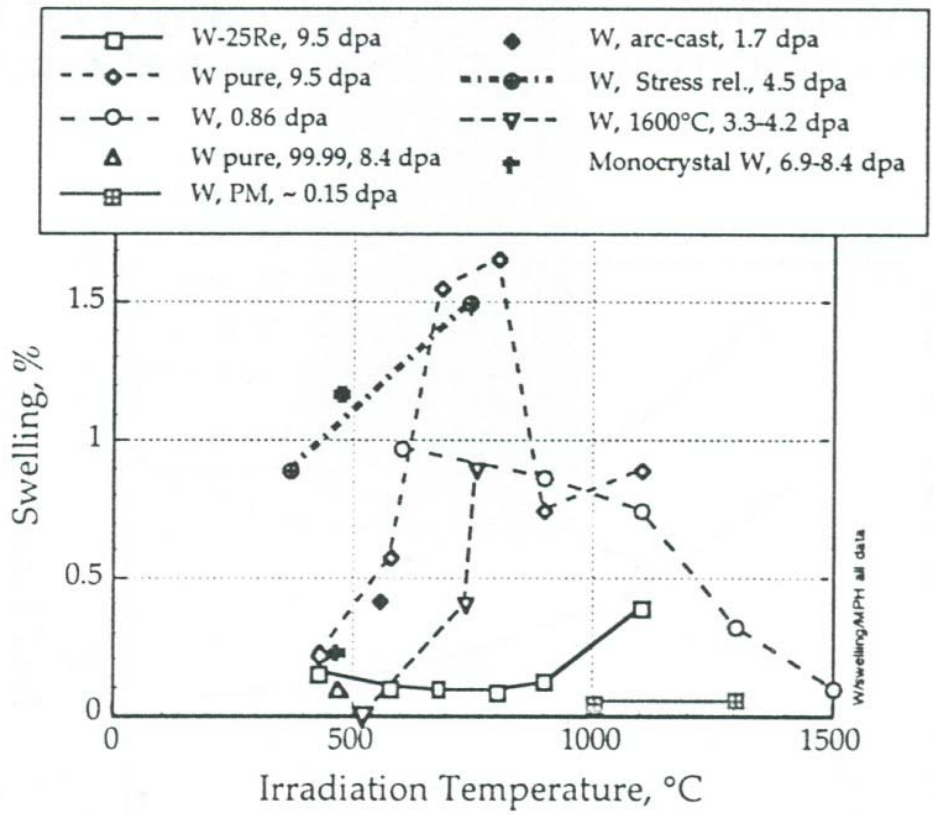

Figure 5. Summary of swelling data for W and W alloys suggesting a peak temperature at ~ 800°C for 5-10 dpa. From the ITER Materials Assessment Report [15].

5. IMPROVING DUCTILITY AND TOUGHNESS: ALLOYS AND MULTI-PHASE MATERIALS

A summary of earlier empirical efforts to improve the ductility of W through alloying and by incorporating oxide or carbide particles may be found in Lassner and Schubert [45]. Currently, several fusion and non-fusion materials programs in Japan and the EU are pursuing several possible routes to developing W-based materials with improved ductility. These are primarily empirical in nature with a focus on W-Re alloys, W-1%La_2O_3, and W-1%TiC. An overview of these efforts was presented by Rieth and Dafferner in 2005 [81].

W-Re alloys

The commercial W-23%Re alloy is now regarded as unsuitable for fusion applications for several reasons:

a) limited data indicate that radiation hardening-induced shifts in DBTT are similar to W,
b) neutron activation issues and enhanced decay afterheat,
c) reduced conductivity compared to W, and
d) the formation of radiation –induced or radiation-enhanced intermetallic phases [82-84].

Alloys containing 0.1-5.0 wt.% Re may have more potential. Based on fracture toughness data, it has been shown that the addition of ~5 wt.% Re reduces the DBTT of W by ~ 300°C; recrystallization and grain growth are also inhibited [85]. The addition of 0.1-1.0% Re has been observed to reduce the DBTT for W single crystals [86]. The results of atomistic modeling of the ductilizing effects of alloying with Re were presented recently by Gludovatz et al. [68] indicating that alloying with Re results in a transition to an asymmetric dislocation core and a significant reduction in the Peierls stress. The possibility that other alloying elements such as Ti, Ta and V could produce similar effects is under investigation.

W-1% La_2O_3

Originally developed as an improved material for welding electrodes, W-1% La_2O_3 is also being considered for high temperature turbines in hydrogen energy systems [87,88]. However there is no materials science basis for expecting improved fracture toughness or improved radiation behavior in the commercially available alloy compared to pure W. The commercially available material contains a coarse distribution of La_2O_3 particles 5-20μ long aligned in the hot-working direction [89], see (Fig 6).

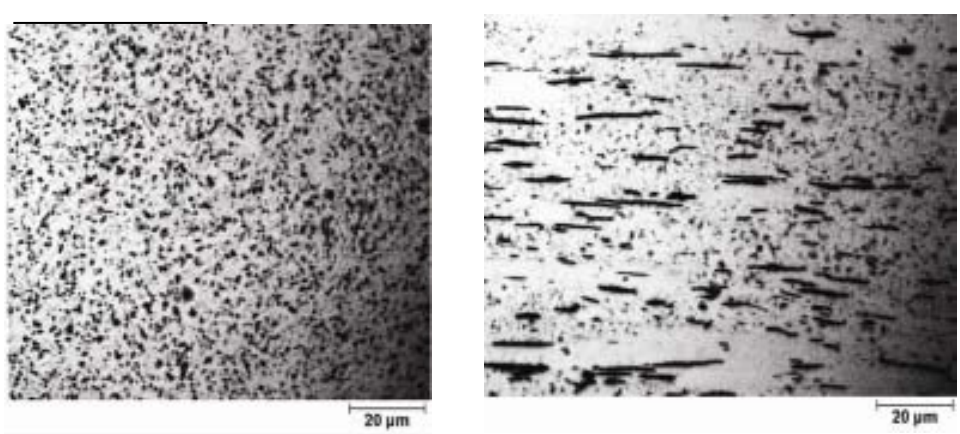

Figure 6. Transverse and longitudinal sections of commercial W- La_2O_3; coarse non-uniform dispersion with La_2O_3 particles strongly elongated in the hot-working direction. From Rieth and Hoffman [89].

A mechanism for the development of the elongated particle morphology during processing has been discussed by Z. Chen et al. [90]. High heat flux testing results indicate enhanced surface erosion related to vaporization of elongated La_2O_3 particles resulting in blistering [91,92]. This material was dropped from consideration for the ITER divertor [15], but it is still regarded as a potential material for the

thimble structure for a He-cooled divertor for DEMO [35, 36] since the La_2O_3 particles appear to inhibit recrystallization to some extent. Efforts to improve the performance of welding electrodes by varying processing parameters indicate that it may be possible to refine the particle size distribution into the nano-size range [93]. It has not been determined whether or not these radically different microstructures would also translate into improved mechanical behavior.

W-TiC

Researchers in Japan are pursuing various empirical approaches to producing TiC dispersions in W via mechanical alloying followed by HIPing at 1350ºC or high temperature forging [94-98]. Kurishita et al. [95] reported that the W grain size became increasingly refined as the TiC content was increased from 0.3-1.5%; average grain sizes in the 50-100 nm range were achieved when processing was carried out in purified Ar. Grain size refinement was attributed to grain boundary pinning by TiC particles Evidence was presented for the precipitation of TiC during processing although there is very little information on the size and number density of the dispersoid [95]. In subsequent work, the concentrations of Ti, C and Ta were varied in a successful effort to suppress the formation of the W_2C phase during processing. It was found that a W-0.3Ti-0.006C alloy exhibited appreciable ductility in room temperature tensile and three point bend tests with un-notched specimens. However, the ductility appears to be related to the very high aspect ratio grain structure and delamination during fracture [96]. Recently it was reported that W-TiC materials exhibit superplastic behavior at temperatures above ~1500°C for TiC contents > 0.25% [97]. The possible advantages of W-TiC materials in terms of improved radiation performance have also been discussed recently [98]. In related studies it has been shown that nano crystalline TiC is formed during prolonged ball milling of micron –sized powders of Ti and graphite [99,100], possibly indicating a means of improving the homogeneity of TiC dispersions.

W-W_{wire} composites.

Composite tungsten materials containing high volume fractions of various metal oxides and carbides have been investigated extensively [45]; however the thermal conductivity of tungsten is substantially reduced by the introduction of 20-40 vol.% of dispersoid. For fusion applications the incorporation of W wires into a W matrix is a more viable approach in which a fiber/matrix interface is engineered to dissipate the crack energy via interfacial de-bonding and friction. Preliminary results of a proof-of principle study were presented recently by You et al. [101].

6. IMPROVING TOUGHNESS THROUGH GRAIN SIZE ENGINEERING

Advances in processing techniques and computational materials research have made it possible to achieve grain sizes in the ultra-fine grain regime, (UFG, d~100-500 nm) and the nanocrystalline regime, (NC, d <100 nm) for a wide range of metallic materials. The drive and interest in nano-crystalline materials comes from the development of higher strength and /or ductility with increasing strain rate and the possibility of superplastic deformation at lower temperatures. Most of this work has focused on fcc materials and the refinement of the grain size of bcc materials into the UFG/NC regimes is more difficult. A number of reviews of this rapidly growing field are available [102-108]. While there is reason to believe that microstructures in the NC regime would result in some advantages in terms of resistance to radiation damage compared to coarse-grained W, the possible advantages in term of fracture toughness and the high temperature microstructural stability of bulk UFG/NC materials remains to be demonstrated [109]. In this regard, Yang and Yang [110] have discussed the competing mechanisms of grain boundary dominated plastic deformation and grain boundary decohesion which possibly control the DBT in nanostructured metallic systems.

The principles of synthesizing nanostructured materials via mechanical milling and high temperature consolidation have been reviewed for example by Koch [102]. This approach, often referred to as bottom-up processing, has advantages when dealing with brittle materials but requires special precautions with regard to contamination and to the development of defects during the consolidation stages. The alternative top-down approach employs a forged or HIPed powder metallurgical product as a starting point and accesses the UFG and NC regimes via the imposition of very high levels of strain, i.e. severe plastic deformation (SPD) processing. (This distinction is somewhat artificial since mechanical milling also involves the imposition of severe levels of plastic deformation). The various techniques employed in the top- down mode of processing have been reviewed for example by Valiev et al [103]; a comprehensive set of papers dealing with SPD of a broad range of metals and alloys has been presented in a Materials Science Forum [111]. Both approaches are currently being utilized in attempts to induce some level of ductile behavior and to lower the DBTT in tungsten. Refinement of the grain size of W and W-10% Re has been demonstrated by Oda et al. [112] via mechanical milling combined with consolidation utilizing spark plasma sintering. The development of nano-sized grains within the deformed powders was tracked via TEM and Fig. 7 illustrates the equiaxed nature of the ultra-fine grains produced by this approach. Evidence was also presented for the suppression of grain growth of nano-grains in the W-Re alloy related to segregation of Re to the grain boundaries. It has been shown recently by Wang et al. [113] that nanocrystalline tungsten powders produced by ultra-high energy milling have greatly enhanced sintering properties; 97% densification was achieved by pressureless sintering in hydrogen at temperatures as low as 1100°C.

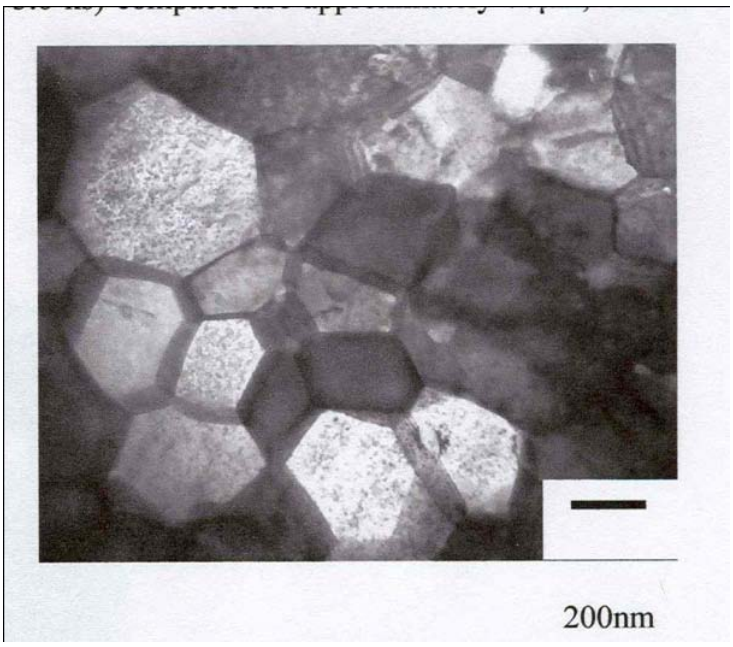

Figure 7. TEM microstructure of nanocrystalline tungsten produced via mechanical milling and spark plasma sintering at 1200°C. From Oda et al. [112].

There is an on-going effort to modify the deformation behavior of W by producing UFG and NC materials with the goal of improving the penetrating power of W-based projectiles [114-116]. Because of difficulties in avoiding rapid grain growth and achieving full density during the consolidation stage during bottom-up processing of pure tungsten, in this area more attention has been focused on top-down processing utilizing Equal Channel Angular Extrusion (ECAP) and high pressure torsion (HPT) [115]. Microstructures of tungsten in the UFG/NC regime produced by ECAP and HPT processing are shown in Fig.8. Under compressive loading conditions at room temperature, it was demonstrated that failure in the UFG materials occurred primarily by localized shearing rather than by brittle fracture [116].

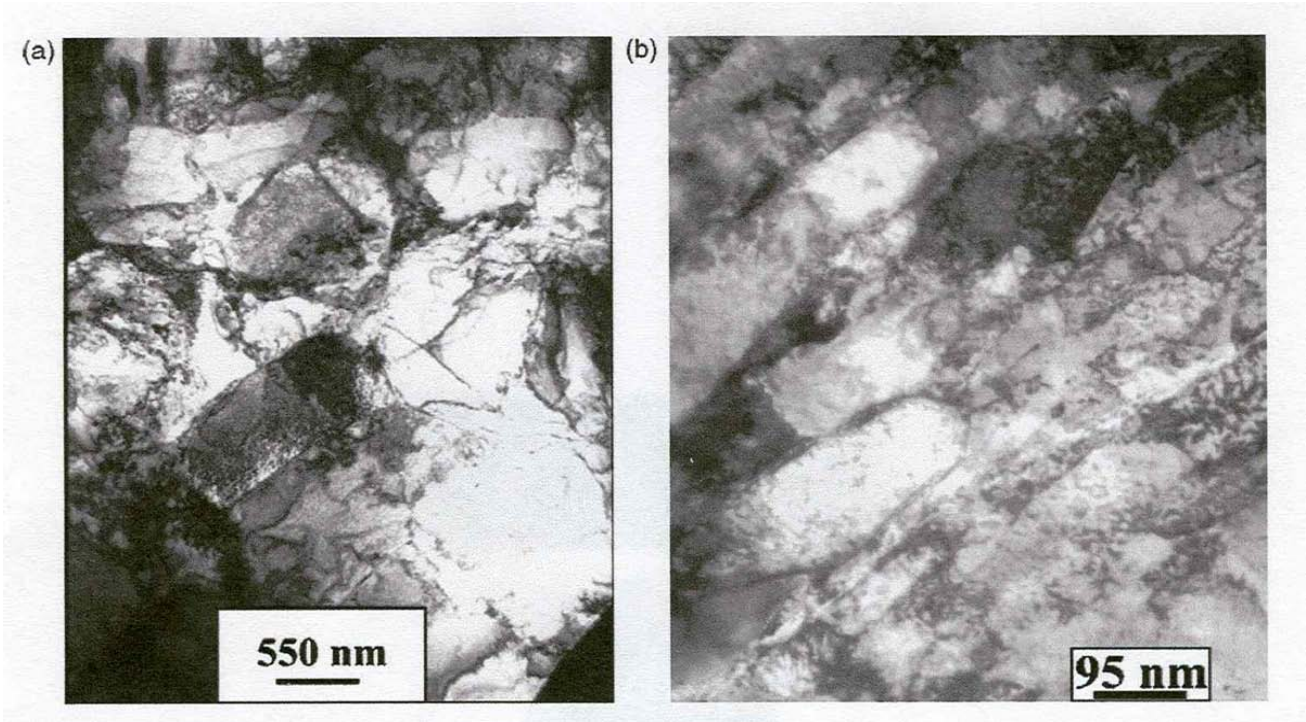

Figure 8. TEM micrograph of pure tungsten (a) processed by Equal Channel Angular Extrusion, four passes at 1000°C followed by lower temperature rolling, average grain size ~500nm and (b) processed by HPT to give an average grain size ~120 nm. Both structures characterized by high dislocation densities and low levels of texture. From Kecskes et al. [115].

Pure W, W-La_2O_3 and K-doped W have also been produced via SPD processing with grain sizes in the ~ 300 nm regime [117]; fracture toughness testing demonstrated a significant increase in RT toughness with increasing levels of deformation and simultaneous reductions in grain size, (Fig.9). Qualitative improvements in the fracture behavior of UFG equiaxed tungsten produced via ECAP were reported by Zhang et al. [118]. Data on the high temperature stability of UFG and NC tungsten is practically non-existent although Zhou et al. [119] recently reported an increased resistance to recrystallization in UFG tungsten fabricated by resistance sintering under ultra-high pressure.

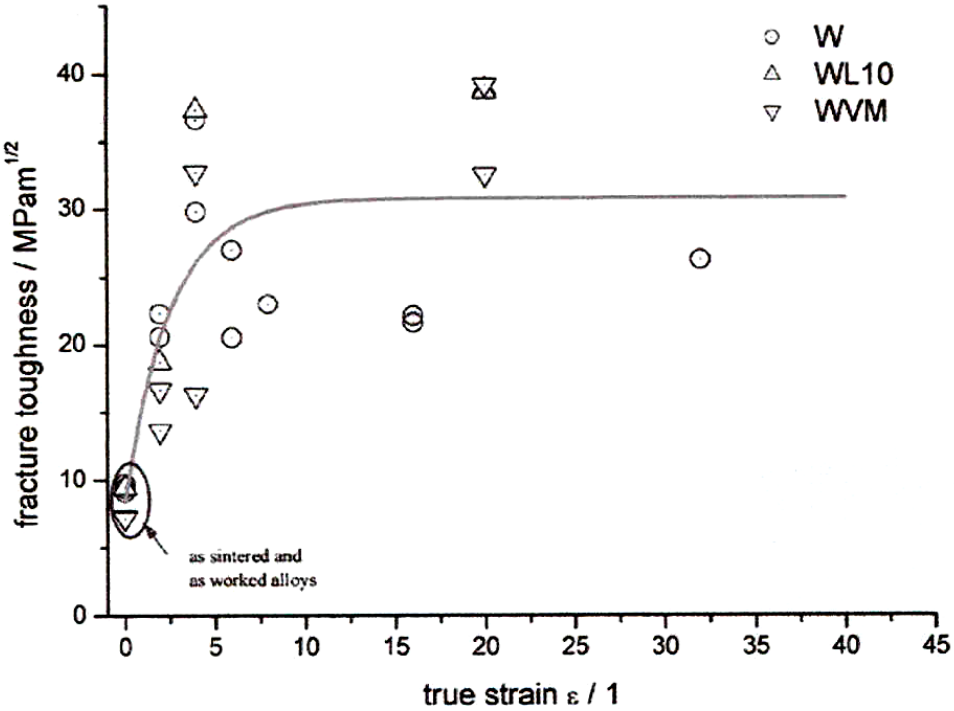

Figure 9. Fracture toughness (DCT specimens) of pure W, W-La_2O_3 and K-doped W as a function of plastic strain applied via high pressure torsion processing; grain sizes ~300 nm. From Faleschini et al. [117].

7. DESIGN OF IMPROVED TUNGSTEN-BASED MATERIALS

Achieving the widest possible operating temperature window for tungsten-based divertor materials presents a major challenge involving not only solutions to the high strength /low ductility paradigm in BCC materials but also the development of strategies to mitigate damaging mechanisms related to plasma –surface interactions, radiation effects and high temperature excursions during off-normal events. A long-term multidisciplinary effort will be needed characterized by close interactions between the materials and PMI communities together with industrial partnerships in order to access a variety of processing technologies.

On-going efforts to improve the ductility of W-based materials based on W-La_20_3, W-TiC and grain size engineering, as summarized above, are unlikely to succeed in producing materials that meet the full range of desirable characteristics in terms of fabricability, improved ductility over a wide temperature range, radiation damage resistance and microstructural resistance to high temperature excursions. Clearly, the development of materials with improved properties in all of these areas simultaneously will require a science-based approach to developing a set of compositional and microstructural design principles, the establishment of microstructure-property relationships, and the appropriate processing-microstructure relationships. To support these efforts, it will also be necessary to develop processing technologies which will deliver the required control of impurity levels coupled with predictable, consistent, and uniform microstructures. In order to develop science-based strategies for improving the properties of W-based materials, the fundamental knowledge –base will need to be strengthened in many areas including deformation and fracture, chemical and mechanical alloying,

formation and stability of carbide and oxide dispersions and solute clusters, high temperature microstructural stability and radiation damage. As noted above, a significant amount of new information is currently being generated in both fusion and non-fusion materials programs in the EU, China, Japan and the US. There is a broad range of theoretical and experimental topics which could be used as a basis for new initiatives within the US Fusion community; these are summarized below in four main categories.

7.1 Critical analysis of the existing data base

The existing literature data needs to be evaluated and analyzed in order to clarify the current state of knowledge in the following areas:

a) The current experimental data base on the fracture behavior of tungsten based materials.
b) The conditions which control transitions from transgranular to intergranular fracture the factors causing de-lamination; the effect of interstitial impurities, grain size and texture; the influence of processing methods and parameters on microstructure and fracture behavior; and the methodology of fracture testing.
c) The relative effectiveness of various approaches to achieving dispersions of oxide and carbide particles and varying particle size distribution thermodynamics of alloy systems, alloying behavior with potential dispersoid-forming elements; mechanisms of mechanical alloying in tungsten
d) Improving ductility and toughness via reduction of grain size into the UFG and NC regimes; examination of the data for FCC and BCC materials to elucidate the potential advantages of grain size engineering as a viable approach to improving the fracture properties of tungsten.
e) Data on the recovery and recrystallization behavior of refractory metals and controlling mechanisms; role of initial grain size and texture, dislocation density, time and temperature.

7.2 Fundamental studies of flow and fracture in tungsten

Theory and modeling work coupled with fundamental experiments is needed in the following areas, some of which are the subject of on-going studies in the EU and in Japan;

a) Definition of principles for microstructural design for higher toughness in low, medium and high temperature regimes; influence of grain size and texture, particle dispersions, dislocation substructures and grain boundary structure.
b) Atomistics of brittle fracture, both transgranular and intergranular; crack tip plasticity in tungsten, dislocation generation and mobility; influence of interstitial and substitutional solutes.
c) Deformation and fracture mechanisms in BCC materials with grain sizes in the UFG and NC regimes; thermal stability of nano-engineered grain structures.

7.3 Theory and modeling radiation damage in tungsten

Nanoscale microstructures designed for improved ductility and toughness may alter the basic radiation damage response of tungsten-based materials and could possibly result in increased recombination rates of point defects and enhanced trapping of helium. Additional needed studies could include:

a) Development of principles for the design of damage resistant microstructures; effects of grain size reduction into the NC regime; dislocation substructures; oxide and carbide nano- scale precipitates as traps; development of solute nanoclusters and their thermal stability.

b) Primary damage, defect production and cluster formation in W migration and trapping of helium and tritium, interaction of dislocations with defects, radiation hardening mechanisms, effect of grain size from micron to nano-scale regimes.
c) Radiation–induced segregation and phase stability in dilute W alloys; potential sources of non-hardening embrittlement.
d) Carefully controlled low dose neutron irradiation experiments to map out the dose-temperature characteristics of radiation hardening, deformation and fracture behavior in polycrystalline and single crystal W

7.4 Alloying and nanoscale strategies developed in other metallic systems

Several new strategies for modifying the strength and ductility of metallic systems and for modifying radiation response have evolved in recent years. The potential application of these approaches to tungsten-based systems needs to be examined in terms of how these new ideas could be utilized in the design of new alloys and microstructures to meet alloy development goals.

a) First principles density functional theory calculations have been used to predict Poisson ratio in Cr alloys and to identify potential ductilizing additives and the design of new materials using quantum mechanical calculations [120].
b) Design principles for nano-clustered alloys and related fabrication technologies have been successfully developed for the NC ferritic steels [121,122]; these alloying principles could possibly be extended to W-based systems in order to develop nanoscale precipitates and solute clusters.
c) The potential for optimizing strength and ductility in a wide range of materials by engineering coherent internal boundaries at the nanoscale has been reviewed recently [123]. Although primarily focused on FCCs, this methodology could possibly provide a new approach to the development of W-based materials with nanoscale sub-structures
d) Evidence has been presented for a reduction in the Peierls stress in W by alloying with Re; it is proposed that this could be the primary source of the observed ductilizing effect of Re; other solutes may have similar effects and so contribute to enhanced crack-tip ductility, Gludavatz et al. [68]
e) Nanocrystalline microstructures via bottom-up and top-down technologies have been developed for a wide range of metallic systems including tungsten [114-119]. Modeling efforts are needed in the areas of the mechanical behavior, thermal stability and irradiation behavior of UFG and NC tungsten-based systems.

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