PSI Experiments in CPP-IPR High Heat Flux (HHF) Device, the Growth of W Microcrystals from Melt Tungsten

3.1 Introduction

To simulate the extreme heat load expected on the plasma facing components of future fusion devices, numerous heat sources have been used including electron, ion, neutral beams, IR heaters or plasma assisted sources [86]. The electron sources allow very good control over the deposited power and uniformity of heat deposition through rastering of the beam. On the other hand, a plasma-based source may have the
The advantage of mimicking the physical/chemical environment of fusion plasma more closely, in terms of ion composition, plasma density/temperature and particle/heat fluxes. It becomes especially crucial for ITER where heat/particle fluxes in the divertor will be of order $\sim 10 \text{ MWm}^{-2}$ and $\sim 10^{24} \text{ m}^{-2}\text{s}^{-1}$, respectively. In this paper, we are reporting the development of a simple, low-cost plasma-assisted heat source, which may be utilized for high heat flux testing of materials under ITER relevant conditions. A segmented torch was used as the plasma source, which operates in a similar fashion as the cascaded arc source in the PILOT-PSI or MAGNUM-PSI devices [87]. This plasma torch configuration was introduced by Maecker in 1960’s, which was later optimized further and thoroughly explored at Eindhoven University of Technology [54,55]. Atmospheric pressure thermal plasmas usually interact with matter in a violent, uncontrolled fashion; hence to achieve better control in terms of depositing heat uniformly the plasma beam is first made to expand in a linear chamber maintained at lower ambient pressure. Achieving such a profile is also critical for the development of plasma assisted techniques for rapid surface engineering of metal cutting tools or vacuum plasma coating processes [88].

Because of its combination of superior properties, such as high melting point, hardness, thermal conductivity, high sputtering threshold and negligible tritium retention, tungsten is considered to be one of the most promising materials for plasma facing components of future fusion devices [18]. However, serious uncertainties still remain regarding the prolonged interaction of the intense fusion plasma with the tungsten plasma facing materials, especially in the divertor region. Accidental events, disruptions or ELMs may lead to uncontrolled deposition of large plasma heat loads and subsequent melting of tungsten [89]. This may lead to further concerns like
drastic changes in surface properties, the release of dust particles in the fusing plasma, reduction in material lifetime or capacity of withstanding high heat flux. It is therefore essential to study the melting processes of this strategically important material under steady state as well as transient heat load conditions. This paper also investigates melting and surface modifications of tungsten exposed to very high heat fluxes and temperatures. A substantial amount of observations exists in the literature on melting of tungsten, either from experiments in tokamaks [90,91] or from plasma surface interaction experiments carried out mostly using transient plasma pulses produced by plasma guns [92,93]. More recently, De Temmerman et al. have explored melting behaviour of tungsten in a linear plasma device, under steady state heat load conditions or in the presence of ELM-like transients [94,95]. Interaction of high power beams with materials is significant also from a material synthesis point of view; as such configurations often lead to highly non-equilibrium conditions in the interaction region, promoting novel nano/micrometer level structures [96]. Previous studies under a controlled surface temperature of the target material have demonstrated the evolution of extremely interesting surface structures, almost up to the nanometer level resolution [97,98]. Surface nano-structuring of tungsten may lead to enhanced resistance to radiation damage in a fusion environment, which is another motivation behind this study [99].
3.2 Experimental details

Fig. 3.1 Schematic of the segmented plasma torch assisted High Heat Flux device.

The High Heat Flux (HHF) device consists of a segmented plasma torch coupled to a double-walled stainless steel linear vacuum chamber (70 cm length, 40 cm diameter), connected either vertically or in the axial direction (Fig. 3.1). The torch has a thoriated tungsten cathode and a stack of copper ring segments separated by Teflon gaskets and a copper anode, each of which was water-cooled by passing 8 litres per minute (lpm) of water at 2.5 bar pressure. The vacuum chamber was pumped down with an HHV made 1500 lpm rotary vacuum pump, and gas flow rates were controlled through Aalborg made Mass Flow Controllers. A DC thyristorized power supply (200 V/400 A) with a high-frequency igniter was used to operate the plasma source. For the production of plasma, high voltage high-frequency pulses are applied between the cathode and the first ring, which initiates a pilot arc between them. The discharge current flows through a 0.8-ohm resistance, and a potential is dropped across it, which lowers the potential of the first ring. Due to the potential difference
between the first ring and the anode, the arc is transferred to the later. The arc between the cathode and the anode heats up the plasma gas, injected at the cathode ring, ionizing a part of it and produces a stabilized, axis symmetric plasma flame coming out from the opening of the anode ring segment. The heat flux deposited on an external target plate was measured with a simple 2 cm diameter stainless steel made calorimeter, placed at a distance of 14 cm from the torch anode nozzle, perpendicular to the laminar plasma beam. While connected to a closed loop chilled water circuit, at steady state, the cold water passing through the calorimeter removes the entire heat deposited on the same. The heat deposited on the calorimeter was estimated by measuring the temperature difference between the incoming and outgoing water (PT100) and the corresponding water flow rate.

![Fig. 3.2 The geometry of Abel Inversion measurements. The plasma jet is assumed to have a circular periphery with radius R, only those portions of the pipe calorimeter which collects plasma heat are depicted here, in grey colour.](image)

The radial distribution of heat flux was also measured by the techniques of Abel Inversion [67,100]. For this, another calorimeter consisting of a copper pipe of 3 mm diameter was used. The calorimeter pipe was placed at different locations across the cross-section of the plasma beam with a spatial resolution of 1.25 mm (Fig. 3.2).
This gives the heat deposited along the line of sight at different positions \( I(y) \), which is related to the local heat density \( F(r) \) as,

\[
I(y) = \int F(r) \, dx
\]  

(3.1)

By inverse formula, we get,

\[
F(r) = -\frac{1}{\pi} \int_R^R \frac{I'(y)}{\sqrt{(y^2-r^2)}} \, dy
\]  

(3.2)

This is the Abel integral equation, through which the heat deposited along the radial distance from the centre was computed. The plasma parameters at the position of the tungsten target were measured by the techniques of optical emission spectroscopy, with a McPherson 1.33 m spectrometer (1800 grooves/mm) and CCD camera (ANDOR DU940P, 2048 × 512 pixels). Integrated light over the line-of-sight passing through the cross-section of the plasma at a distance 12 cm away from the nozzle was collected by a fibre optic cable. The fibre optic cable was placed on a side port of the cylindrical chamber. The plasma temperature was estimated using the Boltzmann Plot method by measuring the following Ar-I lines: 545.1 nm, 549.5 nm, 555.8 nm, 557.2 nm, 560.6 nm and 565.0 nm. The electron density was estimated by measuring the Stark broadened spectral lines of H\(_\beta\) with fitting Lorentzian profiles to the observed spectra. The hydrogen spectral emission was obtained by adding little hydrogen to the Argon plasma. For the high heat flux exposure experiments, the torch is connected vertically to the vacuum chamber, so that there is no motion of the tungsten melt because of gravity. A target holder is placed at a distance of 14 cm from the torch nozzle, on which PLANSEE tungsten targets (99.97% pure, 2.5 cm square sides, 0.3 cm thick) are placed horizontally and grounded, in the line of the plasma beam. The W piece was exposed to pure argon (300A, 25 lpm argon) plasma beam, the pressure
in the vacuum chamber maintained at 20 mbar. To study the surface morphology changes, the entire exposed target plate was placed inside a Scanning Electron Microscope (JSM6360, JEOL), without disturbing the particles connected loosely to the centre of the plate. This experiment was repeated while operating the torch with argon/helium mixtures (300 A, argon 20 lpm, helium 10 lpm) and also with externally biasing the target at -50 volts. Micrometer-level crystals structures were then processed for TEM sample by extracting the crystals from the surface of the tungsten target. The process was initiated by embedding them in epoxy and forcing the epoxy into a 3-mm diameter brass tube and cured in a hot plate at 130 °C prior to curing the epoxy. The tube and epoxy are then sectioned into disks with a wire saw. A disc is then mechanically thinned, dimpled and finally, Ar ion milled at 3 kV followed by 1.2 keV cleaning process to make it electron transparent TEM sample for this study. An FEI, Tecnai G² F30, S-Twin microscope equipped with Gatan Orius CCD camera was used for TEM study. The attached scanning unit and HAADF detector from Fischione (Model 3000) was also used to perform the high angle annular dark-field scanning transmission electron microscopy (STEM-HAADF). The crystallography was studied with X-ray diffraction (XRD, Philips PW 1710, using CuKα radiation). EDX connected to the SEM and Selected Area Electron Diffraction (SAED) connected to the TEM was also used for further characterization of the samples. A simple estimation of the final equilibrium surface temperature of the target was made using the software ANSYS. The calorimetry measured heat flux deposited by the plasma beam under the conditions of the material exposure experiments was the heat input, whereas the only loss was through radiation by the target surfaces. The calculated
values still represent an upper estimate of the temperature, as conduction losses through the tightening nuts were ignored.

3.3 Results and discussion

3.3.1 Development of a High Heat Flux device for material testing

The argon plasma jet from the torch anode expands supersonically into the low-pressure vacuum chamber. During this experiment, the torch was connected horizontally to the chamber, so the jet was moving parallel to the axis of the chamber. Expansion continues until a shock is produced, after which the jet propagates with an almost constant cross-section, in a stable, laminar flow pattern. The position of the shock depends on the pressure in the vacuum chamber and moves away from the torch exit as pressure is reduced [101]. Hence the cross-section or the size of the plasma beam may be controlled through the ambient pressure in the vacuum chamber; at 20 mbar pressure, the argon plasma jet in this system had a visible diameter of about 3 cm. At 15 lpm argon at the cathode, the length of the laminar, collimated plasma jet was measured to be around 10 cm for 100 A plasma current, which increased to 45 and 55 cm for higher currents of 150 and 200 A respectively. The length of the plasma beam increased more rapidly for 20 lpm argon; from 35 cm at 100 A, to 55 cm at 150 A. These results are in agreement with studies on similar plasma torch designs [102]. This may be termed as an ideal configuration for a controlled plasma-assisted heat source, because the laminar plasma could be made to interact with a material target placed remotely even on the other end of the vacuum chamber, increasing the available space for diagnostics.
Fig. 3.3 (a) Heat flux deposited on the calorimeter for different plasma input power and argon gas flow rates, for the 9-ring torch, 20 mbar chamber pressure, (b) Abel inverted radial heat flux profile for 20 lpm argon flow, for three different plasma currents.

The plasma heat load deposited on the calorimeter could be enhanced by merely increasing the input power to the plasma torch system. As the source power supply had limited current capacity (400 A) advantage was taken of the relatively high output voltage (200 V), and input power to the torch was increased further by increasing the number of floating plates between the cathode and anode. This led to an enhanced input voltage, which is explained by the Paschen’s law: for constant pressure, discharge voltage may increase as the effective distance between the cathode and anode is increased [103]. Fig. 3.3a shows the measured heat flux deposited by the 9-ring torch on a 20 mm calorimeter for different plasma powers corresponding to plasma currents of 150, 200, 250, 300 and 350 A respectively, for three different argon flow rates or 15, 20 and 25 lpm. 10 MW/m² heat flux is achieved for 25 lpm argon, 43.7 kW input power (350 A plasma current). The heat delivered by the plasma beam was contributed by electrons, ions and neutral atoms, all together. For every electron-ion pair in the plasma beam arriving on the calorimeter, an energy equivalent
to argon ionization potential is transferred to the surface upon their recombination, while their kinetic energy is also fully absorbed. The degree of ionization on the beam front varies between 1-4% under present experimental conditions; hence neutrals highly outnumber ions, and a large part of the heat flux to the calorimeter is contributed by the hot argon atoms. Fig. 3.3b depicts Abel inverted radial heat flux profile of the argon (20 lpm) operated plasma jet for different plasma currents of 150, 250 and 350 A. This shows that under low-pressure operation, the plasma heat flux actually spreads out over few cms diameter area, which should ensure controlled interaction of the power beam with a material target. A Full Width at Half Maximum (FWHM) may be defined for the plasma beam with respect to the measured heat flux which is about 14 mm at 350 A.

![Fig. 3.3b Abel inverted radial heat flux profile of the argon (20 lpm) operated plasma jet.](image)

Fig. 3.3b Abel inverted radial heat flux profile of the argon (20 lpm) operated plasma jet for different plasma currents of 150, 250 and 350 A.

Fig. 3.4 Measured plasma density with different plasma current and argon flow rates, for the 9-ring torch, at 20 mbar chamber pressure.

Plasma density was seen increasing with argon flow rates as well as plasma current, although somewhat saturating for higher current values (Fig. 3.4). At 25 lpm argon, 300 A plasma current, the plasma temperature was measured to be 0.2 eV,
showing negligible variation with the current. High heat flux exposure experiments were later conducted under this experimental condition only. This corresponds to a plasma heat flux of 9.3 MW/m².

3.3.2 Exposure of tungsten in the High Heat Flux device

Fig. 3.5 (a) The geometry of tungsten target exposure to the collimated plasma beam, (b) photograph of exposed tungsten target, large micrometer particles were seen where the plasma beam had striked the target, and (c) SEM photograph of typical melting-resolidification patterns seen on the exposed target.

A tungsten target was exposed to the argon plasma beam (25 lpm argon, 300 A, 9.3 MW/m² incident flux) for two different durations of 5 and 30 minutes. Through the viewports, the target tungsten surface was seen to get white-hot within just a few tens of seconds after starting the plasma exposure. After taking out the exposed samples from the vacuum chamber, even with unaided eyes, one could see large grains growing on an almost circular area where the plasma beam had interacted with the
material surface (Fig. 3.5b). The target was next examined under an SEM, which shows that the entire top surface of the plate was getting highly corrugated (Fig. 3.5c), and large micrometer granular structures are depositing in the central exposed region. All those morphologies are very typical of re-solidified materials, which confirm that the surface temperature was high enough to lead to melting of tungsten. The outer part of the target surface showed iridescent colours, while the central exposed part was darker in comparison. At an incident heat flux of 9.3 MW/m² under the present exposure condition, ANSYS analysis calculated the maximum surface temperature of the tungsten target to be ~4300 °C after five minutes of exposure, higher than the melting point of tungsten. This temperature may lead to appreciable tungsten evaporation also, although we failed to detect any tungsten emission lines in the plasma spectrum collected from the target interaction region. However, nanometric particles were seen depositing around the interaction region, which must have condensed from this vapour only.

SEM micrograph of the centre of the 5 minutes exposed tungsten sample show a dense coverage with hopper/skeletal crystals, around 20-40 micrometers in sizes (Fig. 3.6a). On the other hand, about 5-10 micrometer polyhedral particles are seen depositing in the peripheral areas, surrounding the primary high heat-flux plasma-material interaction region (Fig. 3.6b). EDX and XRD confirmed those materials to be crystalline α-tungsten only. It is well-known that the final morphology of a growing crystal depends upon the balance between kinetic and thermodynamic control; hopper morphology may be produced when the former dominates [104]. Increase in viscosity of a melt during high undercooling may lead to slower
diffusion/delivery of atoms to the crystal faces as opposed to the corners, which is known to produce this pitted morphology.

Fig. 3.6 (a) Hopper crystals forms on the tungsten target plasma interaction region after 5 minutes of exposure, inset shows crystals with higher magnification, (b) polyhedral tungsten particles growing on the target peripheral region, (c) columnar hierarchical hopper microstructures after 30 minutes exposure, seen from top, and (d) the previous grains photographed from side.

Thermal plasmas with their typically very high working temperature often provide very high super-cooling. Experimental measurements are available which confirm that viscosity of an under-cooled tungsten melt increases as the temperature is reduced below the melting point, which could have led to the formation of those especial crystal structures during this experiment [105]. SEM photographs of targets exposed for longer periods (30 minutes) show crystals growing in size, almost one-dimensional vertical structures growing from a narrow tip like base, of about 200-300
µm in length and 100 µm in diameter (Fig. 3.6c). They also have hierarchical hopper like morphology, which possibly formed from the aggregation of the smaller hopper/skeletal crystals over time. Hierarchical skeletal structures have been identified before in a plasma fusion device, graphitic particles in the Tokamak T-10, by Kolbasov et al. Skeletal structures, have been observed before for other various materials, such as in case of halites forming in salt lakes, complex PbTe crystals synthesized through mild solution method or bismuth metallic Hoppers nucleated from a melt [106,107]. Calcium carbonate, a vital biomaterial with rich polymorphism may also acquire hopper morphology in Calcite form [104].

Fig. 3.7 (a) TEM image of spherulitic polycrystalline structures with nanometer level grain substructure, (b) SAED pattern from red circled area of (a), (c) STEM-HAADF image showing open spherulites (arrow points to the center of an individual spherulite), and (d) SEM image showing dumbbell spherulites (pointed with an arrow).
The exact crystal morphology depends on the undercooling the melt may be undergoing during the condensation process. Shtukenberg et al. had correlated progression of typical morphologies with increasing super-saturation as follows: polyhedral single crystals → skeletal/hopper crystals → spherulitic structures [108]. The term spherulite is used in a broader sense of densely branched, polycrystalline solidification pattern [109]. In this experiment, polyhedral tungsten particles must have formed because of a relatively lower under-cooling the tungsten melt might have undergone in the outer part of the tungsten target. Spherulitic structures were also identified in this experiment through cross-sectional TEM of some particles scraped off from the exposed tungsten target. Spherulites can form in general by the process of growth front nucleation when new crystal grains nucleate at the surface of the parent with a little different orientation than the previous, which eventually leads to the splaying growth at larger length and time scales [109]. This so-called non-crystallographic branching is distinct from equilibrium crystallographic branching, for example as in snowflakes where all branches are still part of a single crystal register only, whereas spherulites are polycrystalline in nature [106]. Cross-sectional TEM photograph demonstrates that the material had sheaf-like radiating overall morphology composed of individual grains with sizes in the range of few to several tens of nanometers, which fits into the classical description of spherulites (Fig. 3.7a). SAED (shown in Fig. 3.7b) was taken from about two-micrometer diameter area of the image shown in Fig. 3.7a, which shows that the material is made of nanometric crystals. This was further supported by the XRD peak broadening of the same sample, from which the average effective Scherrer crystallite size was calculated to be about 30 nanometers. Calcium carbonate polymorphs (vaterite and aragonite) also were
demonstrated to possess substructures in the nanometer regime, which also were explained to have formed by similar non-crystallographic branching process only [110]. Some more SEM images of typical spherulite structures identified in the same sample are presented here, showing a relatively open morphology which is less space filling/less compact (Fig. 3.7c) or a dumbbell type spherulite (Fig. 3.7d). In their seminal paper, Keith and Padden had explained fibrillation of spherulitic crystallites from a melt through growth front of instability, induced by the so-called constitutional super-cooling where impurities play the key role. Shtukenberg et al. also had demonstrated that most of the spherulite forming substances was impure, but foreign inclusions even in the ppm level were sufficient to induce these structures, whereas the basic material (PLANSEE tungsten, 99.97% purity) here already had impurities in much excess of that. Experiments were done with an argon/helium mixture (2:1 ratio) or with a negative bias voltage on the target produced almost similar structures, which indicate that the simple heating only was responsible for the formation of the exotic crystal structures and contribution from specific ion/atom chemistry may be neglected here. However, no such hierarchical re-solidified crystal structures were observed during previous tungsten melting experiments [94]. That may be explained on the basis of the apparent differences in the actual experimental conditions under which they were performed; the presence of the J×B forces in the former could have been responsible for not allowing the formation of those special morphologies. Additionally, the chamber pressure was much higher in the present experiment (~200 times), which also could have favoured the growth of such structures.
Fig. 3.8 Engineering drawing of CPP-IPR Magnetised Plasma Experiment for Plasma Surface Interaction (CIMPLE-PSI) device, two pairs of roots vacuum pumps are employed to produce $10^{-2}$ mbar level pressure in the interaction region, three sets of water-cooled copper coils will produce maximum 0.45 Tesla magnetic field for plasma confinement.

This laboratory had upgraded the existing high heat flux system, first by introducing an axial confining magnetic field for the production of a pure helium/hydrogen plasma beam (in the present experiment the beam gets diffused when operated with hydrogen alone) and further reduction of pressure by two orders of magnitudes. That system should recreate conditions similar to that in the divertor region of ITER like devices and would be most suitable for plasma-material interaction experiments of relevance to the fusion community. Developed in the line of PILOT-PSI or MAGNUM-PSI devices, the CPP-IPR Magnetised Plasma Experiment for Plasma Surface Interaction or CIMPLE-PSI (Fig. 3.8), is now in active state and plasma surface interaction studies of tungsten are being carried out currently. The details about the design, development, commissioning, and
characterization of this system and its subsequent utilization to address PSI studies of tungsten are given in chapter 6 of this thesis.

3.4 Conclusion

A simple, low-cost, segmented plasma torch assisted ITER level heat source (10 MWm\(^{-2}\)) has been developed successfully, which may be used for material testing under extreme conditions. A laminar argon plasma beam of around 3 cm visible diameter was produced at a pressure of 20 mbar, which may be conveniently made to interact with a remotely placed material target. Calorimetric measurements of heat flux and their Abel inversion confirmed that the radial profile widens when the pressure is decreased, which should ensure controlled interaction of the high power beam with a target. The FWHM of the beam was measured as 14 mm for 350 A plasma current. The high density, low-temperature plasma was characterized for plasma density and temperature by optical emission spectroscopic techniques.

A PLANSEE tungsten target was exposed to a high power pure argon beam, which had resulted in surface melting and re-solidification into micron-sized skeletal crystals in the central exposed area of the target. During high under-cooling, increase in viscosity of the metal, leading to limited supply of atoms to the crystal surfaces compared to the corners, is understood to have resulted in these especial crystal morphologies. Relatively lower under-cooling, on the other hand, had grown equilibrium polyhedral particle morphologies on the periphery of the target surface. Upon prolonged exposure, the skeletal crystals aggregate together to produce almost one-dimensional vertical grains of several hundreds of micrometers in length. Spherulites were also observed, with typical radiating, polycrystalline grains in
nanometer size level, which were understood to have formed through non-crystallographic branching process during most intense under-cooling of the tungsten melt. Skeletal particles have been already identified in Tokamak devices, and the present results may be relevant in understanding complex tungsten microstructures that may be forming under extreme heat flux conditions. Complex hierarchical structures are rarely seen in functional materials, which is observed for the first time for tungsten in this experiment [107]. This experiment may also provide a new approach to growing hollow, high surface area metallic microstructures with complex hierarchy, which have been highlighted as most ideal for many environmental applications, for example towards water remediation, bio-sensing, environmental gas sensing, catalytic gas treatment, etcetera. [111]. It may also be used as a controlled model system for a better understanding of the intricate bio-mineralization processes producing similar complex hierarchical natural structures with extraordinary mechanical properties, which scientists are eager to replicate during synthesis processes [112].