A STUDY OF RESIDUAL STRESSES IN VACUUM PLASMA SPRAYED TUNGSTEN COATINGS

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Abstract. Thick tungsten (W) coatings are potential plasma facing materials in fusion reactors, but the coefficient of thermal expansion (CTE) mismatch between W and Cu or steel substrates leads to large thermally induced stresses and premature failure. While inter-layers can be used to grade and distribute stresses away from a discrete planar W-steel interface, in the present study an alternative approach was used based on patterning of the interface with repeating millimetre-scale 3D features or “sculptures”. Up to 2mm thick W coatings were manufactured directly on water-cooled patterned substrates using vacuum plasma spraying (VPS), without any inter-layers. Synchrotron-based white beam, high energy X-ray diffraction measurements of lattice parameters was used to obtain maps of residual strains and stresses in slices through the VPS W coatings. Residual elastic lattice strains were deduced from energy-dispersive diffraction profiles collected by two detectors mounted in the horizontal and vertical diffraction planes, providing information about lattice strains in two nearly perpendicular directions lying in the plane of the coating. On the basis of these data, maps of residual stresses in the VPS coatings were constructed. The findings are discussed in the context of the geometry of the substrate-coating interface and any inelastic processes operating to relieve and manage successfully the stresses induced by the thermal expansion mismatch.

Keywords: residual stress, synchrotron X-ray diffraction, vacuum plasma spraying

Introduction

Application of coatings and layered composite structures on metal parts allows components to withstand intense heat and abrasion. However the process of manufacturing coatings and layered composites most commonly involves high temperature gradients and intensive heat transfer between the different components of the system. The process therefore produces significant residual stresses which can lead to distortion, cracking, and coating delamination. It is the combination of temperature gradients and different thermo-physical properties of the materials involved that leads to the formation of thermal stresses in the composite at different stages of the manufacturing process. The final state of residual stresses affects the structural and functional properties of the coating as well as the component reliability during operation. Therefore, residual stress analysis is an important tool for the optimisation of coatings and layer composite manufacturing processes [1-4].

It is well known in the fusion community that thick tungsten (W) coatings on steel substrates are a promising material for plasma-facing components (PFCs) because they offer a combination of good thermal conductivity and high sputtering thresholds, therefore manifesting very low erosion rates during ion bombardment. Nevertheless, difficulties remain in the processing or machining of W for engineering components because generally W is too brittle and too dense to form structural components on its own [5-6]. In addition, the mismatch in CTE between W and steel, typically 4.5 and 12.5 x $10^{-6}K^{-1}$ respectively, leads to the generation of large thermally induced stresses near the interface during thermal cycling. Producing a through thickness graded coating involving mixed W/Steel inter-layers can successfully re-distribute thermal stresses away from a single planar interface to a number of convoluted W/steel interfaces, increasing lifetimes [5,6].

W coatings

In this study, two vacuum plasma sprayed (VPS) coating types with different W morphologies were used; further manufacturing and microstructure details can be found elsewhere [5-6]. Firstly, a W/steel graded coating of about 0.5-0.6mm overall thickness comprising five distinct layers on a 5mm-thick mild steel substrate was considered. The graded coatings were produced in the following way: mild steel substrates were first coated with a Diamalloy 1008 (Sulzer Metco, Fe-17Cr-11Mo-3Ni-3Si-3Cu-4B) bondcoat. Then, different W/Diamalloy fractions were applied as intermediate layers onto the bondcoat, increasing the W fraction from 0% to 100% in a step-wise fashion, with a top coat of 100% W. This was achieved using separate Diamalloy and W mass controlled powder injectors in the VPS torch [5,6]. From bondcoat to top coat, the intermediate layers were 75% Diamalloy 25% W, 50% Diamalloy 50% W, and 25% Diamalloy 75% W.

Secondly, thick (~2mm) VPS W coatings were produced directly on a 5mm thick surfi-sculpted 316L stainless steel substrate. Surfi-sculpt is an e-beam based surface manipulation technique developed by TWI, UK that allows a regular pattern of millimetre scale, complex 3D surface "sculptures" to be manufactured [5,6]. A schematic drawing and magnified microstructure of both samples are shown in Figures 1(a) and (b) respectively.

Residual stresses

High energy dispersive synchrotron X-ray diffraction was used to characterise the residual stress states in the VPS W coatings. The specimens were cut through thickness to produce 'slices' about 1mm thick for transmission diffraction investigation. Residual elastic lattice strains were deduced from the diffraction profiles collected by two detectors mounted in the horizontal and vertical diffraction planes, providing information about lattice strains in two nearly perpendicular directions lying in the plane of the slice, i.e. parallel and normal to the coating surface. On the basis of these data, maps of residual stresses in the VPS coating slices were constructed.

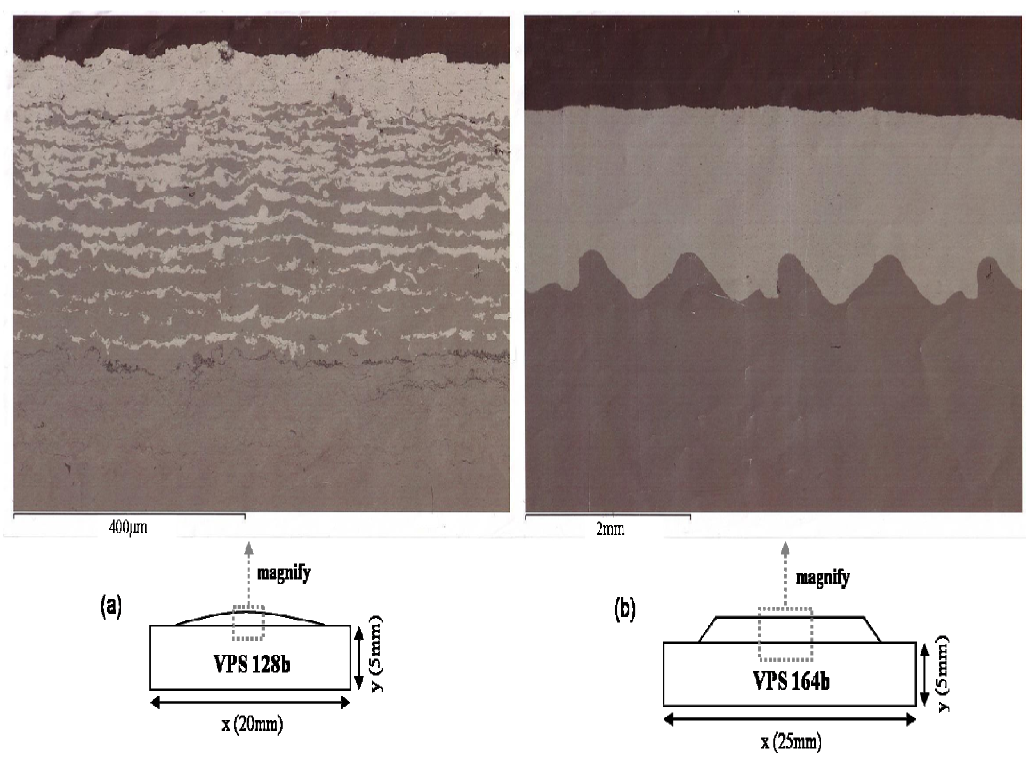

Fig 1. Microstructure and schematic representation of graded (VPS 128b) and surfi-sculpt (VPS164b) W coatings and substrate

Synchrotron X-ray diffraction occupies a special place in the field of residual stress measurement because it can provide information relevant at all structural length scales, and is non-destructive.

It utilises the simple relationship provided by Bragg's law, $n\lambda = 2d^{hkl}\sin\theta$, where n is an integer, λ is the wavelength of the radiation, d^{hkl} is the interplanar distance between planes of Miller indices (hkl), and θ is the angle that the incident beam makes with the lattice planes (called the Bragg angle). Bragg's law provides the relation between the spacing of atomic planes in crystals and the angles of incidence at which these planes produce the most intense reflections.

Diffraction measurements were performed in the white beam energy dispersive mode with twin-detector setup in the high energy synchrotron beamline ID15 at ESRF, Grenoble, France. The white beam mode provides a very high counting efficiency as a broad bandwidth is used to collect multiple reflections simultaneously.

The strain directions, which are parallel (ε_x) and perpendicular (ε_y) to the coating surface, were measured simultaneously. Both detectors were mounted at fixed scattering angle of $2\theta = 5°$. The beam spot size was 0.2mm (x direction) × 0.5mm (y direction) for the graded coating and 0.2mm × 0.2mm for the surfi-sculpt coating. A line scan through the top coat to the bottom edge of the mild steel was collected for the graded coating. A 2D strain mapping (mapped area: 2.4mm×3mm, corresponding to 13points×16points) around the interface was collected for the surfi-sculpt coating.

The obtained data were analysed by single peak fitting [7] and GSAS (General Structure Analysis System) Pawley refinement [8]. In both cases strain error was lower than 10^{-4} which is consistent with the results presented in literature [9-10]. The lattice parameter values obtained in this way were used to calculate residual strain/stress according to procedures described previously [11]. The single peak fitting used a custom written Matlab routine to approximate the experimental data using the either Gaussian or Pseudo-Voigt peak profiles. Each peak is characterised by position, amplitude and peak width FWHM (Full Width Half Maximum); there is no need to input the instrumental parameters and crystal structure information into the program. The results then provide information about the so-called grain group average lattice strain. The grains that belong to this group share the same orientation of hkl lattice plane normal that coincides with the bisector of the incident and scattered beams. However, the interpretation of macroscopic strain on the basis of a single diffraction peak position is susceptible to errors associated with the inhomogeneity of elasto-plastic deformation between grains. Therefore, in addition to fitting individual single peaks, it is possible to perform a Rietveld or Pawley refinement on the entire diffraction pattern collected. If the crystal structure of the material is known, then the positions of the observed lattice reflections can be predicted. Pawley refinement is a similar approach to Rietveld refinement that accommodates the variation in peak intensities by allowing the intensity of individual reflections to vary freely, while the peak positions are determined in the usual manner from the unit cell dimensions. This approach provides an empirical average of the different reflections and has shown to be particularly convenient for the description of the overall deformation of the polycrystal.

Results and discussion

Graded coatings

Figure 2 shows the XRD analysis of an intermediate layer, which has the composition of 50% Diamalloy and 50% W, along the y direction. It indicates the peaks belonging to the W phase and iron phase from Diamalloy and other phases were found. The unknown peaks are likely to come from other minority phases present within Diamalloy or oxides formed during the plasma-spray process. As the cumulative volume fraction of these phases is unlikely to exceed 5%, the effect of minority phases on the overall macroscopic stresses was neglected.

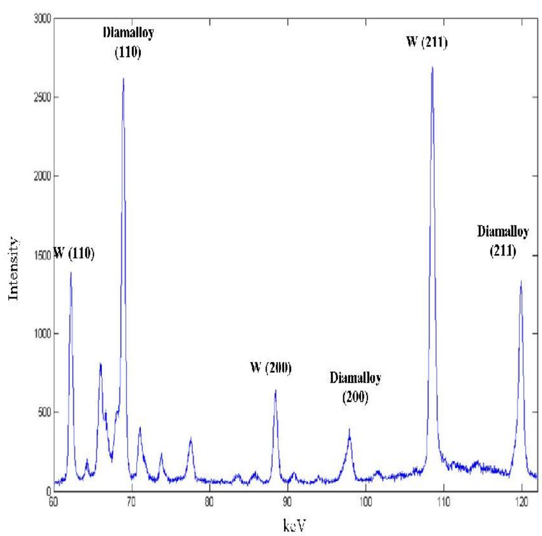

Fig 2. Diffraction pattern of the 50% Diamalloy and 50% W intermediate layer in the graded coating

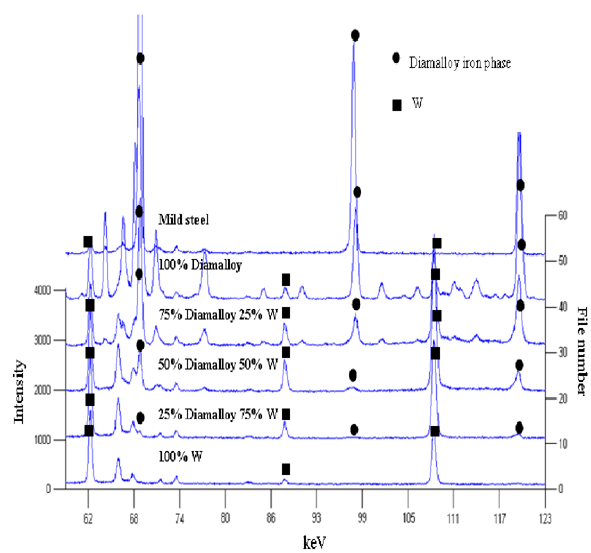

Fig 3. Diffraction patterns from the graded coating

Figure 3 shows the XRD patterns for the cross section scan from the top coat down to the substrate. The intensity of α-Fe peaks reduced proportionally with reducing nominal Diamalloy fraction, whereas the W intensities increased initially and then decreased at the bond coat, and not in agreement with expected monotonic increase with W fraction. Likely reasons include variation in the grain size of W, as shown in the plot of W peak width (related to grain size) in Figure 4, and/or the development/preponderance of a more distinct texture in the rapidly solidified W droplets [5].

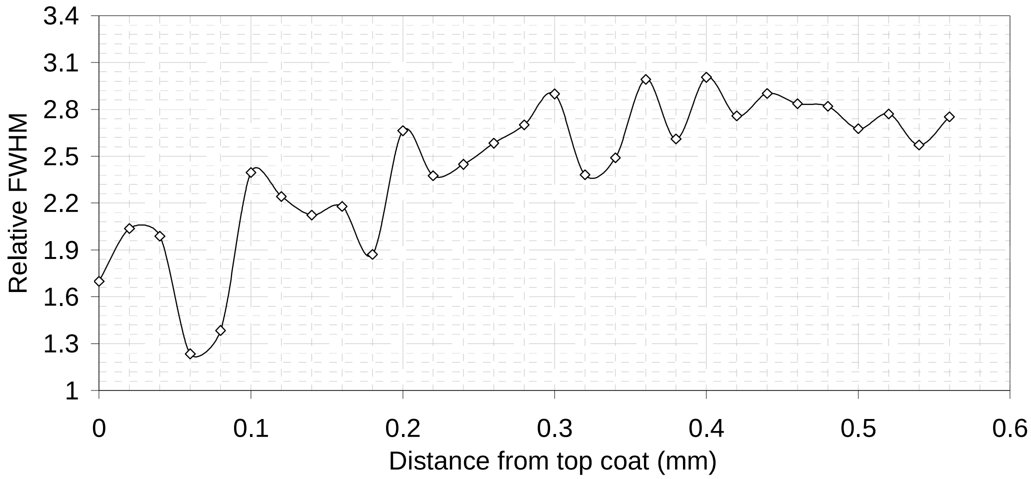

Fig 4. Distribution of peak width from the coating to the substrate of VPS128b

Figure 5 shows the W BCC lattice parameter through the thickness of the graded W coating, corresponding to the strain distribution in the direction parallel to the surface. The strain decreased between the surface and the 50% Diamalloy 50% W intermediate layer, and then stayed relatively constant closer to the bondcoat. Because the fraction of W decreased in this region, the fitting error increased slightly.

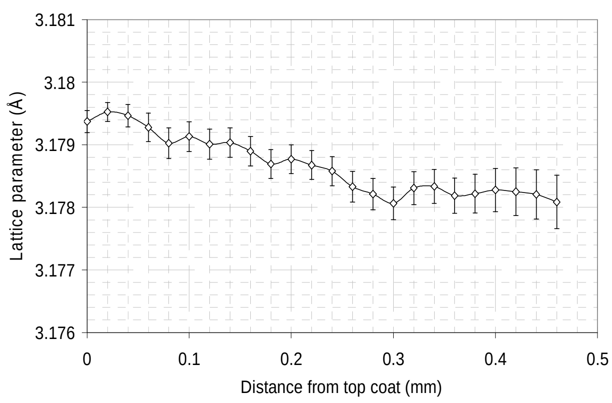

Fig 5. Distribution of the W lattice parameter for the graded W coating

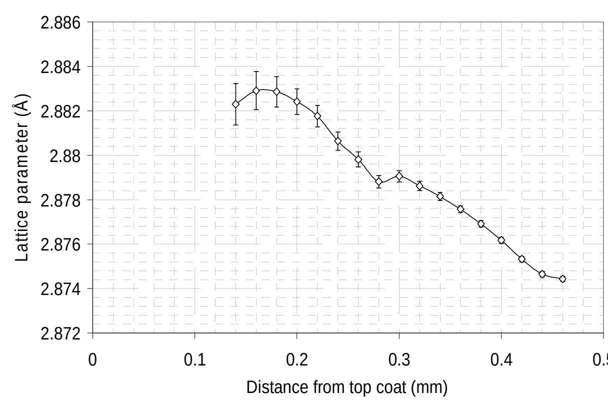

Fig 6. Distribution of α-Fe lattice parameter the graded W coating

Figure 6 shows the α-Fe BCC lattice parameter. Because there was no Diamalloy in the top coat, results are plotted from the 25% Diamalloy 75% W intermediate layer. There was a slightly different slope of lattice parameter with distance beyond the 50% Diamalloy 50% W intermediate layer.

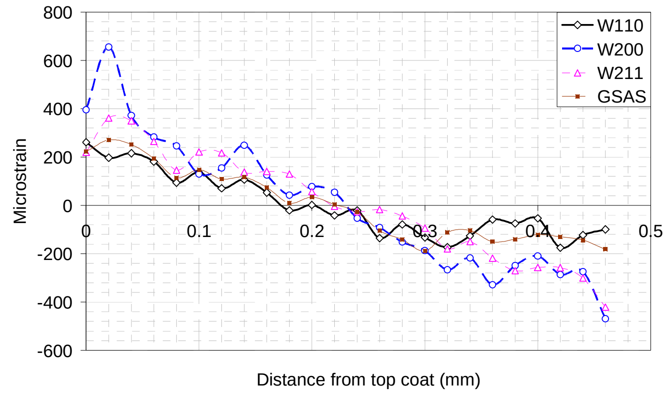

Fig 7. Distribution of residual elastic strains for the graded W coating

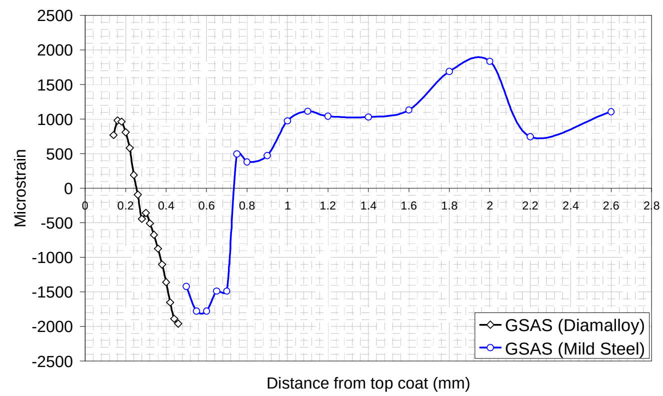

Fig 8. Distribution of residual elastic strains for Diamalloy and mild steel in the graded W coating

The same data are replotted in Figures 7 and 8 in the form of strain-depth profiles for W and Diamalloy components of the coating (as well as the mild steel substrate). The results in Figure 7 show that the strains between single peak fitting data for 110, 200, and 211 reflection of tungsten and GSAS refined data are well agreed. Figure 8 shows the strain distributions of Diamalloy and mild steel, both of which are based on Iron. It is seen that compressive strains exist in the vicinity of the interface, turning to tension out of the region.

The broad conclusion from these plots is that the grading of the steel/W in inter-layers was successful in accommodating material property mismatch, giving a relatively smooth strain variation from the top coat to the substrate, without sharp discontinuities. The experimental results demonstrate that the magnitudes of residual stress in the substrate and the W layer are reduced by about 30% compared to those found in a single layer W coating [5].

<u>Surfisculpt W coatings</u>

The calculated strain data were analysed with the assumption of plane stress, and then the residual stresses (nominal) were calculated using Hooke's law based in the two strain components ε_x and ε_y. Due to the absence of reference strain-free samples, the unstrained lattice parameters for each material were obtained from average values collected along 2 lines at traction-free edges for each material, which were assumed to carry no or little residual macroscopic elastic strain (and hence stress). The Young's modulus used was 411GPa for W and 193GPa for Fe, with Poisson's ratio of 0.28 for W and 0.3 for Fe respectively. The obtained stresses are shown in Figure 9 in the form of contour maps. In the substrate/coating region, transverse stresses were localised at specific regions associated with the surface sculptures, while stresses in the longitudinal direction were distributed more homogeneously along the entire boundary regions.

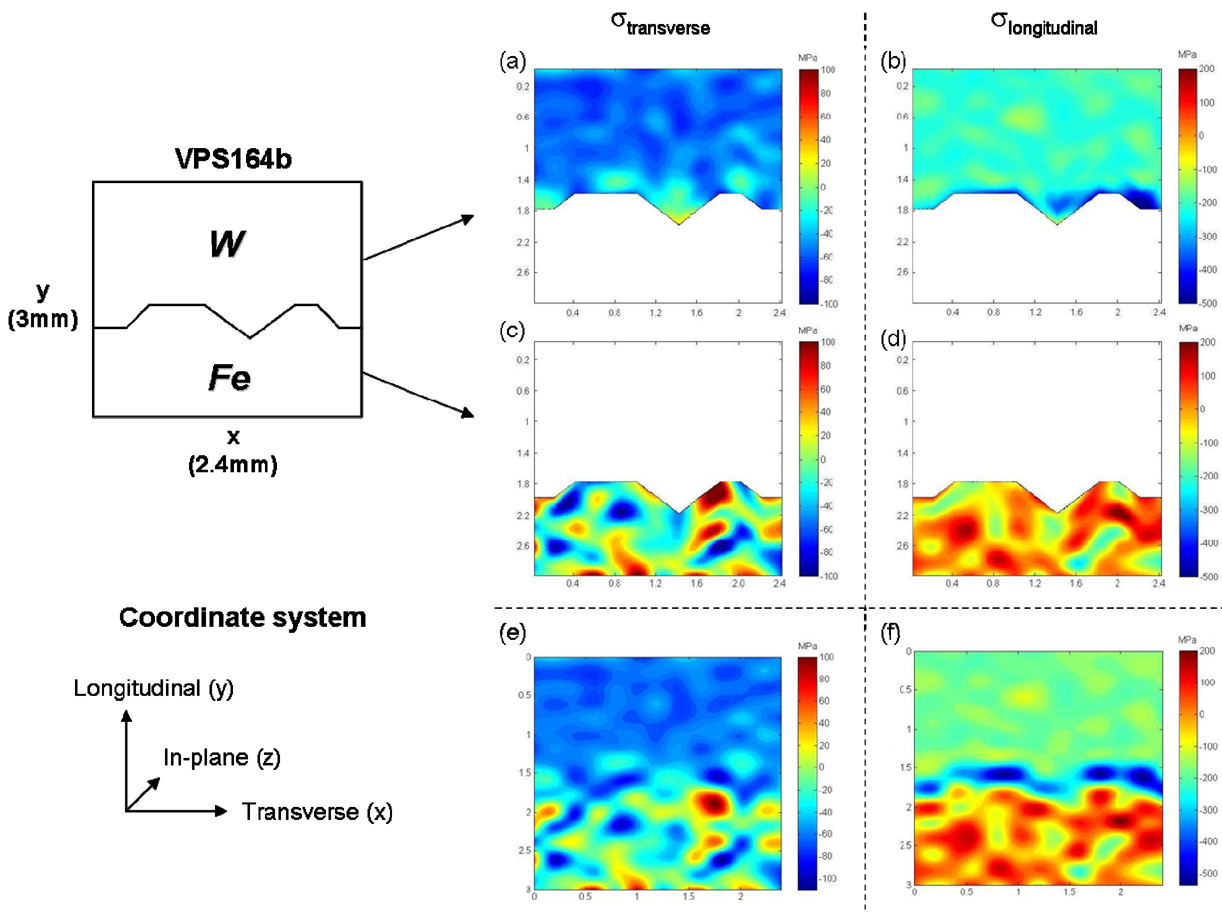

Fig 9. Distribution of mapped area and 2D mapping of residual elastic stresses in the surfisculpt W coating: (a) transverse residual stress in W; (b) longitudinal residual stress in W; (c) transverse residual stress in Fe; (d) longitudinal residual stress in Fe; (e) transverse residual stress in whole sample; (f) longitudinal residual stress in whole sample

Figure 10 shows the distribution of von Mises stress in the surfisculpt W coating, obtained by combining the information on phase-specific strains from Figure 9. It is of importance to study von Mises stress in coated materials since the complex stresses exist in the interface, leading to failure. Most of the W coating contained relatively low von Mises stresses, suggesting elastic CTE strains were relieved in some way and that it may perform well under heat flux service conditions. The substrate experienced relatively large von Mises stresses, while the region of highest von Mises stresses were concentrated in the vicinity of the interface. This finding agrees with the empirical observation of good spallation resistance of surfi-sculpt W coatings. By considering the relatively low stresses in the majority of the W, the drastically improved W resistance to spallation (compared with a simple W on steel planar coating arrangement) can be understood to arise from regulated segmentation (micro-cracking) of the W around stress concentration surface sculptures, while retaining sufficient adhesion to retain the coating under further heat cycles [5]. The surfisculpt arrangement operates as an intermediate solution between a fully bonded coating (that provides excellent thermal contact for cooling but is subject to large residual stresses) and a loosely attached coating (that relieves or “manages” thermal mismatch stresses but reduces thermal efficiency).

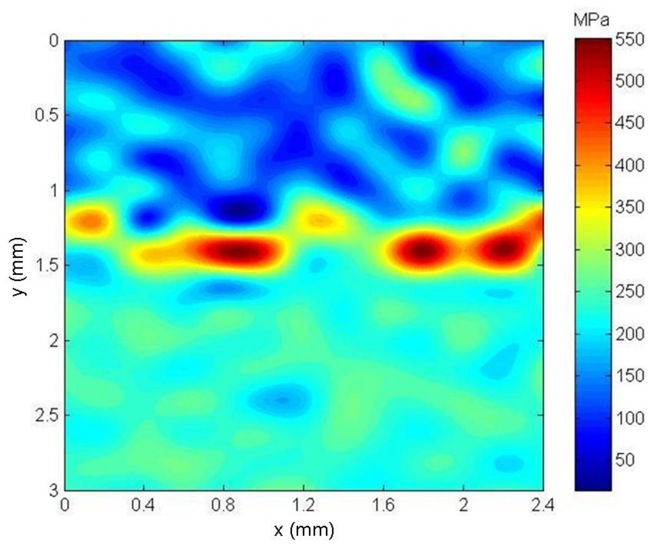

Fig 10. Distribution of von-Mises stress in the surfisculpt W coating

Summary

Thick tungsten coatings are being considered in the fusion community as candidate plasma facing materials due to the useful combination of properties of tungsten, such as high density and melting point that restricts local re-deposition of sputtered atoms and provides a low erosion rate. However, the coefficient of thermal expansion (CTE) mismatch between W and Cu or steel substrates leads to large thermally induced stresses and premature failure. In this study, two different approaches were investigated for manufacturing thick tungsten coatings on steel substrate via vacuum plasma spraying (VPS): graded W/steel inter-layers and a steel substrate surface with regular millimetre-scale sculptures. Residual stress measurements were conducted using the white beam energy dispersive mode with twin-detector setup at the high energy synchrotron beamline ID15 at ESRF, Grenoble. The graded coating produced a relatively smooth residual strain variation across the W/steel inter-layers and successful re-distribution of thermal stresses. The surfisculpt coatings showed that most of the tungsten coating was subject to relatively low von Mises stresses, while the substrate experienced higher von Mises stresses. Residual elastic stresses were concentrated in the vicinity of the material interface and this is suggested to indicate local micro-cracking and segmentation of the W, but there was no evidence of macroscopic coating failure/spallation. The use of surface sculptures or patterns to deliberately regulate segmentation as a way of relieving thermally induced stresses is suggested.

Acknowledgements

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