WPMAT-PR(17) 18630

M Sanchez et al.

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Preprint of Paper to be submitted for publication in
Acta Materialia

This work has been carried out within the framework of the EUROfusion Consortium and has received funding from the Euratom research and training programme 2014-2018 under grant agreement No 633053. The views and opinions expressed herein do not necessarily reflect those of the European Commission.

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Interfacial characterization by TEM and nanoindentation of W-Eurofer brazed joints for the first wall component of the DEMO fusion reactor

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Keywords

Eurofer97, tungsten, fusion reactor, interfacial characterization, plasma facing component

Abstract

The interfacial characterization of W-Eurofer97 brazed joints, which could be implemented in the first wall component of the future DEMO nuclear fusion reactor, has been carried out by TEM and nanoindentation. The Eurofer97-braze interface is characterized by the formation of two reaction layers formed during the melting stage of the filler in the brazing process, which reported high hardness and modulus values. The obtained microstructure supposes a homogenous transition between the base material and the braze. However, in the W-braze interface a more defined interface is formed, where the presence of an intermetallic transition compound has been identified. The combination of both techniques allows to study the resultant microstructure and to analyze the mechanical response of the phases, which constitute the interface and could be helpful to control the brazing conditions avoiding undesirable brazeability problems.

Introduction

The development of joining technologies of some components for the next generation of fusion power plant (DEMO) is one issue which nowadays needs to be implemented. One of these components is the first wall, where dissimilar material such as tungsten and Eurofer97 need to be bonded [1]. Tungsten as sacrificial material will provide high sputtering and temperature resistance due to its adequate thermo-physical properties for the proposed application. Eurofer97, on the other hand, as selected structural steel material for the DEMO reactor, will support the whole component [2].

Due to the different physical properties of the two base materials, heterogeneous bonding procedures are preferred to mitigate the possible stress caused during service operation. In these procedures, both interfaces play and important role in the quality of the joint achieved and, therefore, its characterization could provide important information regarding the presence of undesirable phases or lack of metallic continuity [3, 4].

In this paper the interfacial characterization W-Eurofer97 brazed joints using 80Cu-20Ti as filler material has been carried out. The Eurofer97-braze interface is characterized by the formation of several reaction layers between Eurofer97 and the molten filler, when temperature is above the liquidus one of the filler material, as it was previously described in previous works [5]. However, the small size of these layers makes difficult its characterization by conventional characterization techniques (SEM, DRX) and a more detailed characterization using nano-techniques (TEM and nanoindentation) should be used to study both microstructural and mechanical characterization of the phases that constitute the interfaces [6, 7], which are a key parameter in the quality of the obtained brazed joint.

Experimental procedure

The base materials used for the joint were tungsten (>99.97 %, *Plansee*) and Eurofer97 with the standard composition and microstructure after normalizing and tempering [8]. The fabrication procedure of the filler consisted of laminating a mixture of 80Cu-20Ti powders with polypropylene carbonate as organic binder (powder/binder weight ratio: 95/5) to obtain flexible tapes of 250 μm thickness [9, 10]. The liquidus temperature of the filler measured in previous studies is 885 ºC [10]. Brazing tests were carried out in a high vacuum furnace to avoid oxidation with a residual pressure of 10^{-6} mbar. The brazing cycle consisted of heating up to 960 ºC during 10 min and the heating and cooling rates were 5 ºC /min [9].

The microstructural characterization of the interfaces was carried out by Transmission Electron Microscopy (TEM, *Philips CM200*) operated at 200 kV. TEM sample preparation consisted of the typical lift-out procedure of one lamella from each interface prepared by Focus Ion Beam (FIB) using a *Zeiss Auriga* with a Ga ion beam accelerated from 30 to 2 kV (Figure 1). The lowest energy was used in the last step of lamella fabrication to remove as much as possible the Ga damage on the surface.

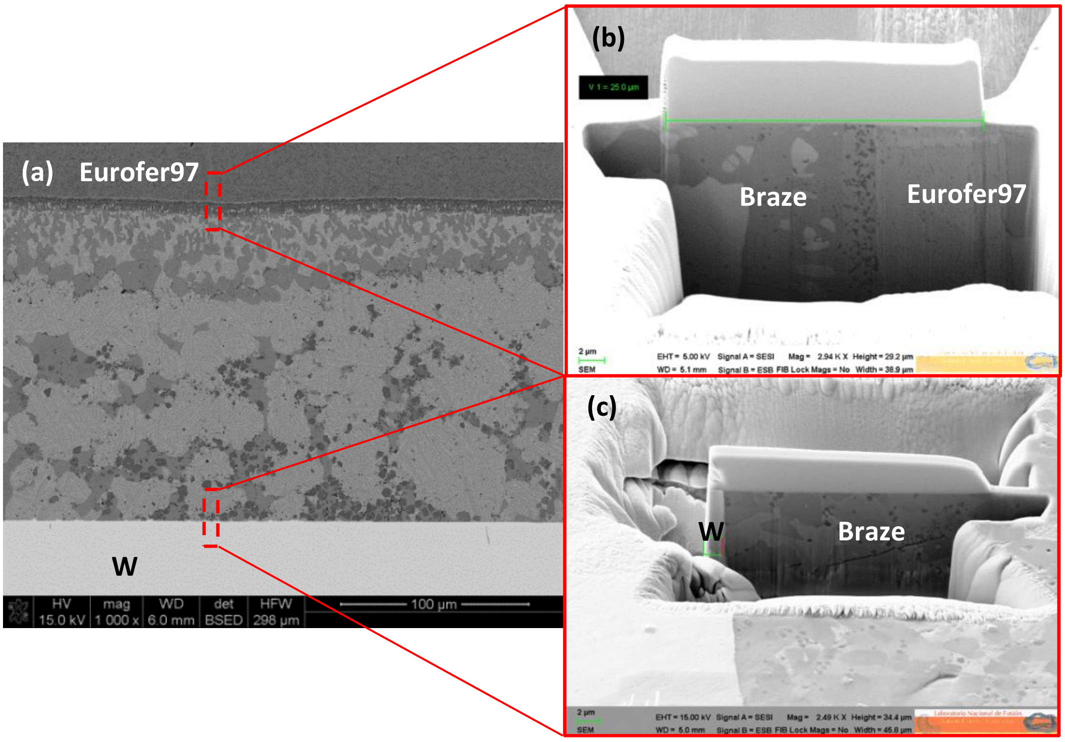

Figure 1. (a) Cross section image obtained by SEM of the W-Eurofer97 brazed joint using 80Cu-20Ti as filler material. (b) and (c) FIB images of the lamellae extraction process of Eurofer-braze and W-braze interfaces, respectively.

The mechanical properties, in terms of hardness and nanoindentation modulus, of the phases, which constituted both interfaces were evaluated by nanoindentation by means of a *MTS Nano Indenter XP* device using two different work modes and a pyramidal Berkovich tip. In the first one, called XP or quasi static, a load of 2 mN was applied. In the second one, called continuous stiffness measurement (CSM), a depth was set (200 nm). The former mode was carried out by applying several indentation lines that crossed both interfaces with a small angle with respect to the interface to ensure that all phases were tested. However, for the latter mode isolated indentations on selected phases were made.

3. Results

3.1 Microstructural characterization of the interfaces

Eurofer97-braze interface

The Eurofer97-braze interface is characterized by the formation of several reaction layers associated to the interaction of the Eurofer97 with the titanium of the filler alloy. This last element is commonly used to enhance the wettability properties of the filler materials due to the formation of low interfacial energy reaction products, which reduce the contact angle of the filler/base material system promoting the wetting and, therefore, filling the joint clearance giving rise to high continuity interfaces.

Figure 2a shows the reconstruction of the Eurofer97-braze interface lamellae created by several TEM images, where the different phases that constitute the interface are clearly observed. The detailed analysis of the first reaction layer (Figure 2b) showed that the microstructure was constituted by a sub-micrometric grain size matrix with high proportion of TiC precipitates homogeneously distributed inside it, identified by electron diffraction (ED) (Figure 2c). The matrix of the studied area had a thickness of 2-3 µm and it was composed by a Fe_2Ti phase (Figure 2d).

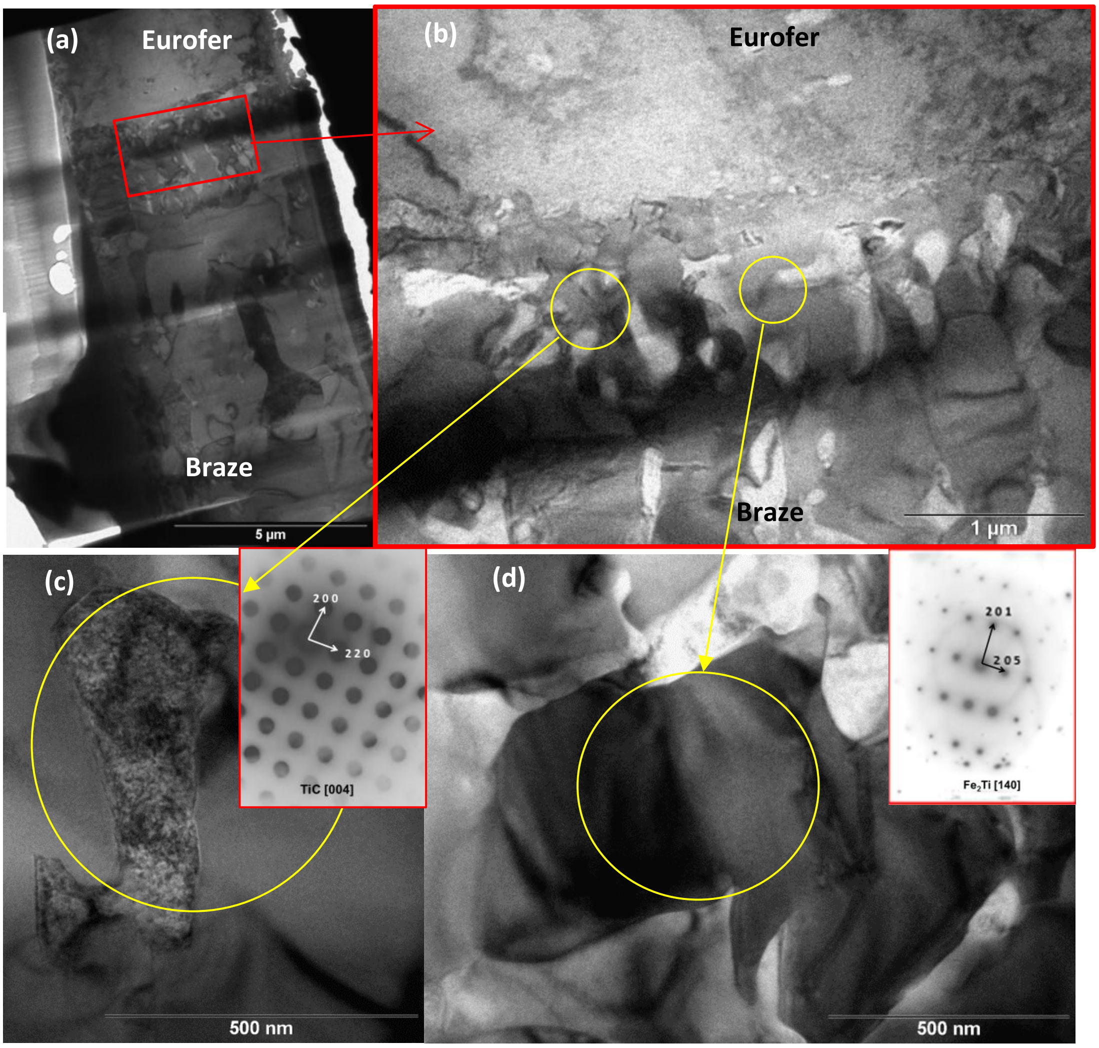

Figure 2. TEM images of the Eurofer-braze interface: (a) reconstruction of the lamellae, (b) detail of the first reaction zone, (c) detail of the TiC precipitate and (d) detail of the matrix.

A second reaction layer was also identified at the interface, and it was characterized by two crystallographic phases with columnar structure (Figure 3a). The columnar shape of the phases could be associated to the temperature gradient formed in the joint during the cooling stage of the brazing cycle. During the cooling stage, the solidification front progressed from the outermost part of the interface (base material was cooler) to the inner part of the braze giving rise to these characteristic microstructures. At this region, copper plays a more important role as main element of the filler material composition. The first phase characterized, identified as A in Figure 3b, was identified as a Cu-Ti-Fe ternary compound $Cu_{0.8}Fe_{0.2}Ti$ and the second studied phase was pure Cu (B in Figure 3b). This pure copper phase comprised the majority part of the braze matrix that extends from the second reaction layer of the Eurofer97-braze interface until reach the W-braze interface. The ductile character of the copper matrix could act as a stress relieving mechanism of the thermal stresses caused during the service life of the component by the mismatch of the thermal expansion coefficient of the two base materials.

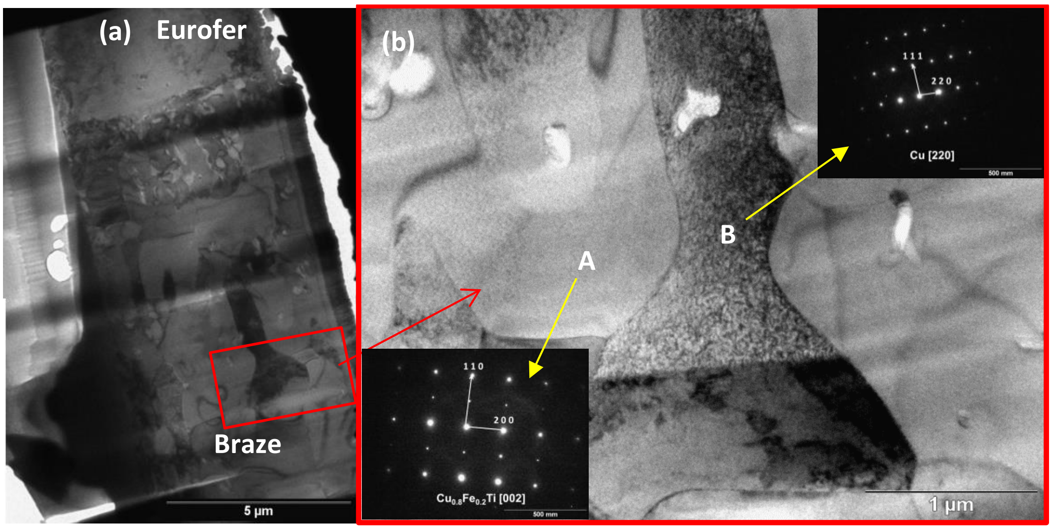

Figure 3. (a) TEM image of the lamella and (b) detail of the second reaction layer of the Eurofer-braze interface.

Tungsten-braze interface

With regard to the W-braze interface, TEM characterization showed the formation of a more defined interface between the base metal and the braze due to the lack of appreciable chemical reactions or diffusion phenomena (Figures 4a and 4b). However, Cu_3Ti phase was identified as intermediate phase between the braze and the base material possibly associated to a higher affinity of this compound to tungsten (Figure 4d). The existing literature regarding Ti and W affinity has demonstrated that Ti and Ti compounds tend to react with tungsten, specially at the grain boundaries, giving rise to partial grain boundary melting [11]. Besides, Ti on the contrary that Cu has certain solubility (25 % at.) into W above 740 ºC according to the equilibrium phase diagram compilated by J.L. Murray [12] and, therefore, its affinity with tungsten is promoted.

According to simulation studies carried out by T. Chehtov et al. the stresses generated during the joining process and, afterwards, during the considerable thermal fatigue associated to reactor operation are located mainly at the W-braze interface [13], which are caused by the mismatch in the Coefficient of Thermal Expansion (CTE) of the two base materials [14]. Therefore, it is expected that a possible crack would propagate following this interface and more specifically following the Cu_3Ti intermetallic compound, compromising the interfacial toughness.

The analysis of the lamella at the braze zone (Figure 4c), showed the presence of a homogenous Cu matrix of the braze, where the presence of TiC was identified. No high density of dislocation was observed, presumably due to the higher temperatures of the brazing, which increase the dislocation mobility.

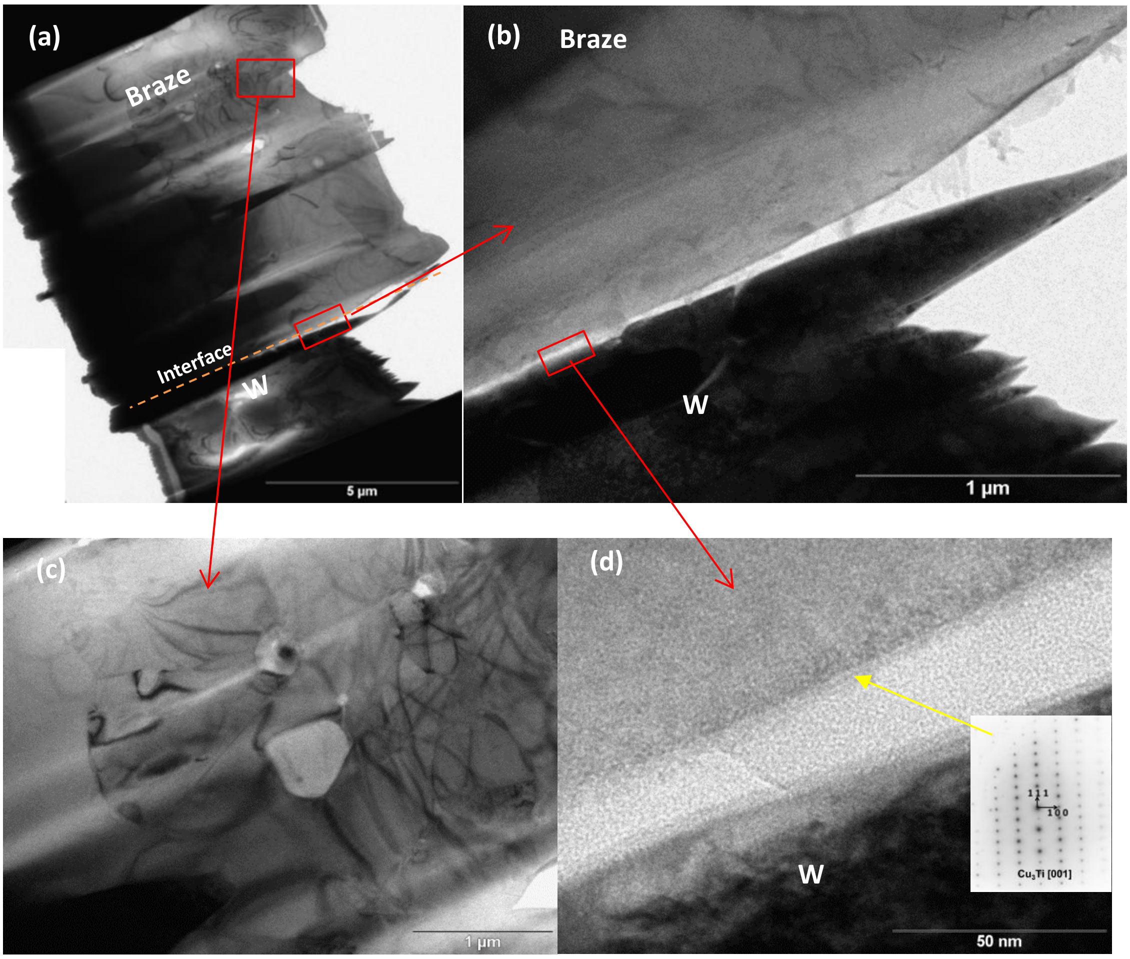

Figure 4. TEM images of the W-braze interface: (a) reconstruction of the lamella, (b)

detail of the interface, (c) detail of braze close to the interface and (d) high magnification image of the W-braze transition phase.

3.2 Mechanical characterization of the interfaces

Eurofer-braze interface

Figure 5 shows the hardness and modulus profile of a line across the Eurofer97-braze interface. The first 5 indentations (white points) correspond to the Eurofer97 base material, whose modulus and specially its hardness presented homogenous values corresponding to a material with homogenous mechanical properties. Afterwards, the following 9 indentations (yellow points) correspond to the first reaction layer formed during the brazing procedure, previously identified by means of ED as intermetallic Fe_2Ti with a high proportion of TiC precipitates inside. Both hardness and modulus values were considerable higher compared to the Eurofer base material. The high dispersion of the data was caused by the heterogeneous microstructure of the layer, depending on the specific location of each indentation the values could be influenced by the different proportion of TiC.

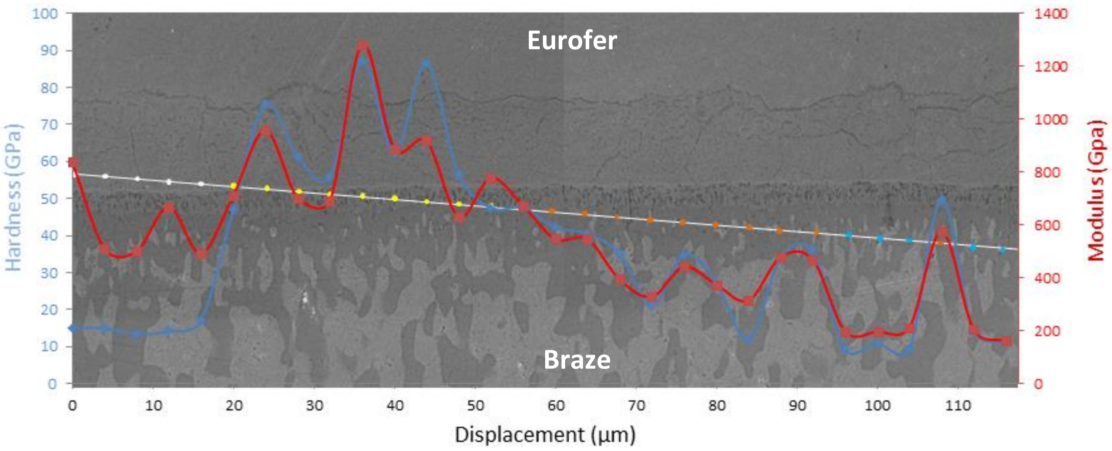

Figure 5. Hardness and modulus profile of the phases that constituted the Eurofer-braze interface.

The microstructure of the second reaction layer, as it was characterized by TEM technique, was constituted by a columnar shape phase of $Cu_{0.8}Fe_{0.2}Ti$ crystallographic phase. The hardness and modulus (orange points) were in the range of the Eurofer97 base material values and were considerably lower than those of the first reaction layer. This could be associated to the high copper proportion of the phase, which provide a certain ductile character to the layer.

Finally, some indentations were made on the Cu matrix between some columnar shape phases (blue points). Its hardness was around 10 GPa showing a ductile behavior comparable to the Eurofer97 base material. As it was previously commented, an intermediate ductile phase between the base materials could be beneficial to relieve the thermal stresses generated during operation life of the component.

The hardness and modulus analyzed by CSM mode indicated that the values obtained from the profile were overestimated due to the influence of the indentation size effect when low loads are applied, as studied in the same material [15]. In the case of the hardest phases (Fe_2Ti and $Cu_{0.8}Fe_{0.2}Ti$) the associated displacement to a load of 2 mN were 45 and 60 nm, respectively (dashed line in Figures 6b and 6c). Therefore, the above reported values are close to the maximum of the load-displacement curves. However, in the case of the more ductile phases (Eurofer97 and Cu) the displacement was higher (80 and 90 nm, respectively) and, therefore, the overestimation in those cases was lower (dashed line in figure 6a and 6d).

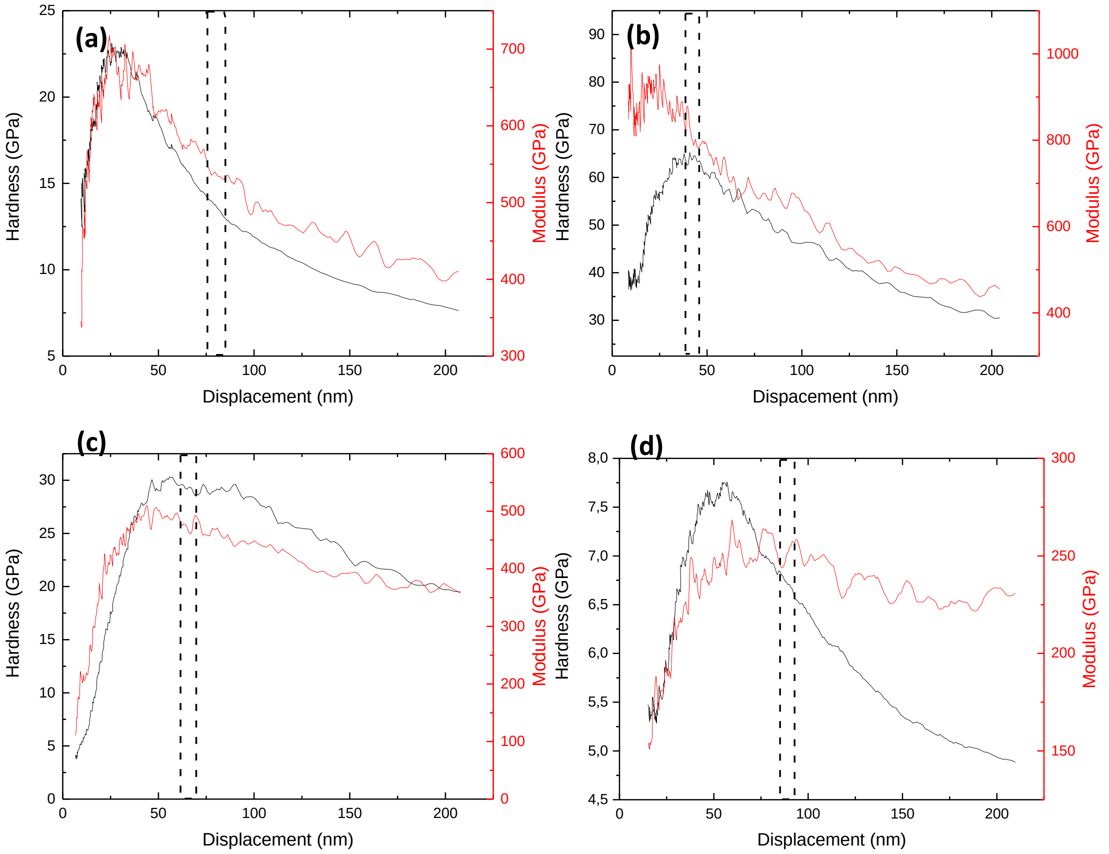

Figure 6. Hardness-displacement and modulus-displacement curves of the Eurofer-braze interface phases: (a) Eurofer, (b) Fe_2Ti, (c) $Cu_{0.8}Fe_{0.2}Ti$ and (d) Cu.

The presence of more rigid phases below the studied one could have a deeper influence on the obtained values, especially in those phases with a low elastic modulus. This effect can be observed in the copper CSM curves where the modulus kept constant as the load or displacement increased. In this case, the modulus values could be influenced by the presence of the $Cu_{0.8}Fe_{0.2}Ti$ phase below the indented copper due to the heterogeneous microstructure of the interface, specially at this zone where the Cu-Ti-Fe ternary phase and copper coexisted.

W-braze interface

The braze close to the W base material was constituted by Cu and a ternary Cu-Ti-Fe compound. The ternary phase, although it was not observed by TEM due to its heterogeneous distribution over the interface, it was identified in previous SEM analysis

[9] and had a higher Ti content with respect to the ternary $Cu_{0.8}Fe_{0.2}Ti$ phase formed at the Eurofer97-braze interface, according to EDX microanalysis. The hardness and modulus of all studied phases are shown in Figure 7.

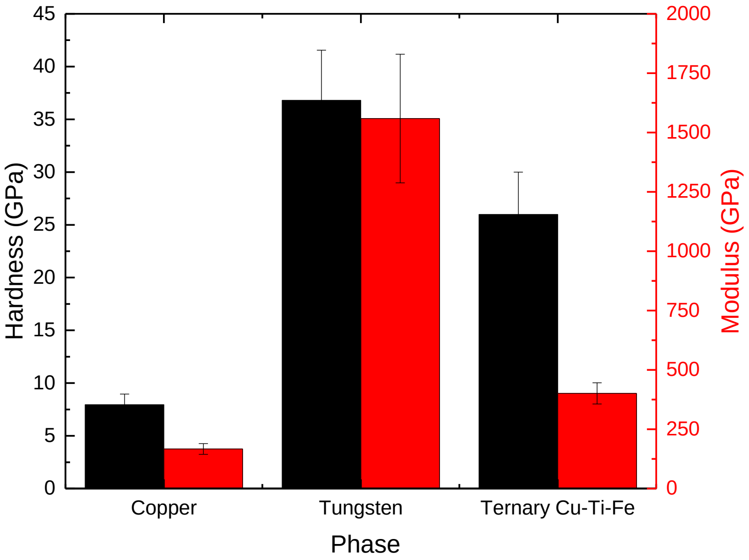

Figure 7. Hardness and young modulus of the phases that constituted the W-braze interface.

Tungsten, as it was expected, is the hardest and most rigid phase followed by the ternary $Cu_{0.8}Fe_{0.2}Ti$ and finally copper. The hardness values of the ternary phase at W-braze interface are similar to those obtained at the Eurofer-braze interface despite the difference in Ti content. Finally, copper shows similar hardness and lower modulus according to the mechanical characteristic of the phase.

The analysis of the CSM curves showed a similar mechanical behavior to that obtained at the Eurofer-braze interface, where the hardest phases such as tungsten and Cu-Ti-Fe ternary phase reported overestimated values (Figures 6a and 6c). The displacement associated to 2 mN load in those cases were 53 and 62 nm, respectively, which corresponded to values close to the maximum of the hardness/modulus-displacement curve. Regarding copper phase, the displacement was 95 nm for 2 mN load and the values were in agreement to those obtained in the Eurofer-braze interface (Figure 6b).

However, the stabilization of the hardness/modulus at higher displacement occurred at larger values possibly caused by the presence of some harder phase below the studied one.

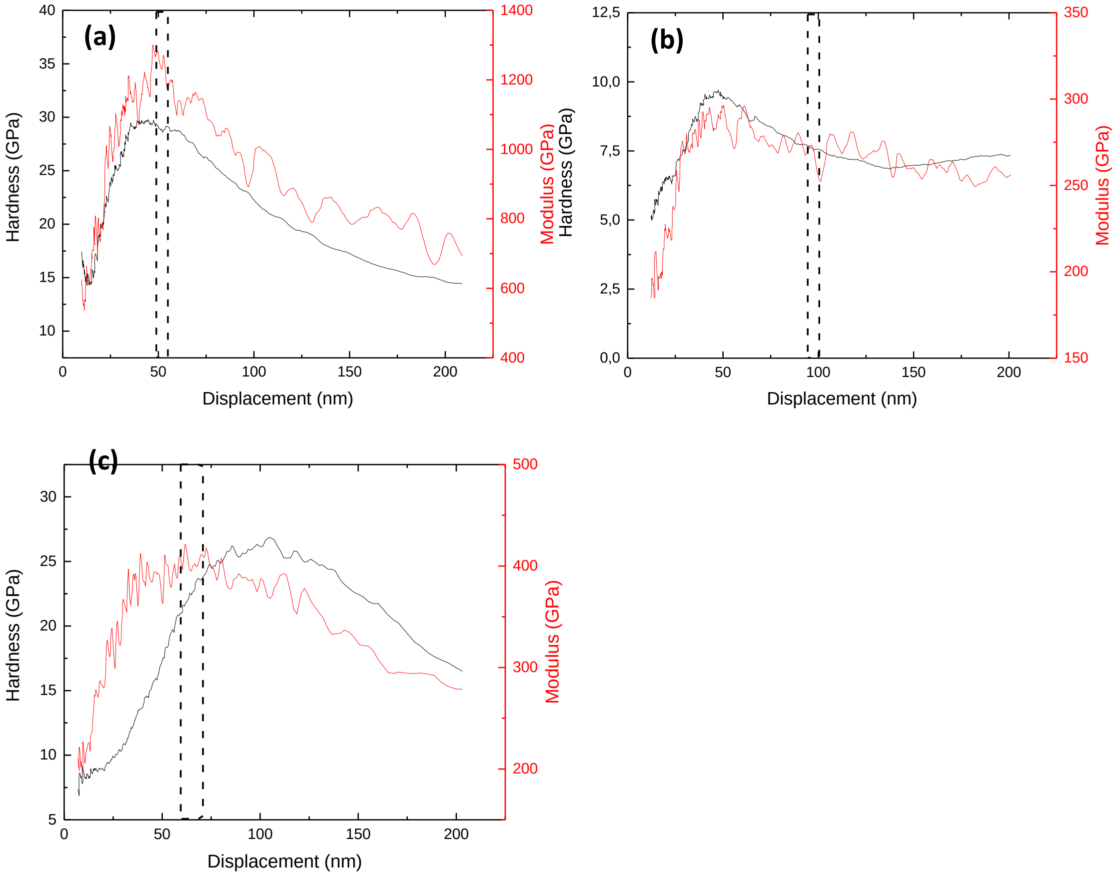

Figure 6. Hardness-displacement and modulus-displacement curves of the W-braze interface phases: (a) Tungsten, (b) Cu and (c) $Cu_{0.8}Fe_{0.2}Ti$.

Although the interface is constituted mainly by copper, as it was demonstrated in the microstructural analysis by TEM, the lack of copper solubility in tungsten and the presence of the Cu_3Ti intermetallic compound could overcome the ductile benefit effect of the copper matrix studied in this section leading to failure of the interface under certain circumstances.

Conclusions

Microstructural and mechanical characterization of the phases that constituted the interfaces of the W-Eurofer brazed joints has been carried out using TEM and nanoindentation techniques.

The filler used was 80Cu-20Ti, which melted at the brazing temperature and reacts with the Eurofer base material giving rise to the formation of several reaction layers. The first one was constituted by a high hardness and modulus phase of Fe_2Ti with an approximately thickness of 2-3 μm. The presence of TiC precipitates inside this layer was detected by TEM electron diffraction. The second reaction layer was formed by two crystallographic phases ($Cu_{0.8}Fe_{0.2}Ti$ and Cu) with lower mechanical properties.

The characterization of the W-braze interface showed the formation of a more defined interface, where the presence of a Cu_3Ti transition phase between tungsten base material and the copper of the braze was identified. The combination of an intermetallic compound and the accumulation of residual stresses at the interface caused by the mismatch in the CTE could explain the fracture mechanism of the brazed joint. At this interface, copper shows a ductile behavior compared to tungsten and could help to extend the service life of the brazed joint under the expected thermal fatigue.

Microstructural and mechanical properties were in complete agreement. Hence, the combination of both nano-techniques allows to obtain important information of one of the most important characteristics of the brazed joints as they are the interfaces and, therefore, to obtain valuable information of its possible behavior during service life such as interface integrity, presence of intermetallic phases with brittle behavior and possible fracture mechanisms.

Acknowledgments

This work has been carried out within the framework of the EUROfusion Consortium and has received funding from the Euratom research and training programme 2014-2018 under grant agreement No 633053. The views and opinions expressed herein do not necessarily reflect those of the European Commission. The authors would like to acknowledge to MULTIMAT-CHALLENGE program (S2013/MIT-2862). The authors also thank Spanish National Fusion Laboratory at CIEMAT, and especially the Fusion Technology Department to give the possibility of using the FIB device.

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