Development of High Strength Tungsten/Oxide Dispersion Strengthened Ferritic Steel Joints by Innovative Thermal Stress Relaxation Technique Based on Phase-Transformation-Induced Creep Deformation

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The realization of fusion reactors rests on the function of the blanket systems with high thermal efficiency consisting of armor and structural materials. The fabrication of the blanket requires upgrade of bonding technology which makes the components reliable. The combination of tungsten and oxide dispersion strengthened ferritic steel (ODS-FS) is considered to be adequate for the application to the fusion divertor components. The issue of diffusion bonding of tungsten with ODS-FS is to reduce thermal stresses caused by a large difference in thermal expansion coefficient between the two materials. In this study, an innovative thermal stress relaxation methodology during cooling has been newly proposed as a key technology in joining, which is based on the phenomenon of $\gamma \rightarrow \alpha$ phase transformation-induced creep deformation of low carbon steel inserted between tungsten and ODS-FS. In the cooling pattern where the joint was slowly cooled at a rate of approximately 1.5°C/min from 800°C to 600°C. The application of the technology to the diffusion bonding of tungsten/ODS-FS resulted in a very high bonding strength of 480 MPa, which has never been achieved so far by the conventional bonding methodologies.

Keywords: diffusion bonding, insert material, thermal stress relaxation, transformation-induced creep

1. Introduction

The dissimilar bonding of tungsten and oxide dispersion strengthening ferritic steel (ODS-FS) has been required for the development of nuclear fusion divertor with high heat removal capability and high resistance to high heat load, since tungsten is well known as a refractory metal with highest melting point, superior resistance to physical spattering, etc.1,2) and ODS-FS is superior in high temperature mechanical properties3,4). Bonding of tungsten and ODS-FS is hence a critical for the fabrication of high-performance blanket, and various bonding processes have been applied for the joint so far.5–7)

A difficulty in bonding tungsten with ODS-FS is that a large difference in the coefficient of thermal expansion (CTE) between the materials causes a rather large thermal stress at the bonding interface so that the joint was damaged by cracking at tungsten near the interface during cooling after bonding. According to the previous study on the estimation of thermal stress by finite element method (FEM)8), a tungsten/ODSS joint generates an equivalent stress more than 1.5 GPa in tungsten during cooling to ambient temperature after diffusion bonding at 1250°C. In order to reduce the thermal stress, an insertion of material between tungsten and ODS-FS is often considered to be effective because of stress relaxation at the insert material. The insert material should also meet the requirements such as reduced radio-activation, high melting point as ODS-FS and the absence of brittle phase formation by chemical reactions between tungsten and ODS-FS.

In our previous research9), an oxide dispersion strengthened vanadium (ODS-V) alloy was selected as an insert material that was a reduced activation material and had a thermal conductivity and CTE of which the values were between those of tungsten and ODS-FS. However, the tungsten/ODS-V/ODS-FS joints were easily broken in tungsten, indicating that the ODS-V insert did not sufficiently reduce the thermal stresses. Therefore, pure iron insert, which is more malleable, was used in the next step, and it was shown that the pure iron was effective to increase bonding strength. Since cracks in the W/ODS-FS joints were always observed in the tungsten close to the joint interface, the bonding strength of the joints was sensitive to the fracture toughness of tungsten that depended on grain structures such as grain size, grain shape and grain orientation9).

Phase transformation-induced creep (PTC)10–13) is known as a transformation super plasticity phenomenon which can be applied to thermal stress relaxation in the insert material during cooling after bonding. The phase transformation generates dislocations around the new phases according to the difference in the specific volume between the two phases13). The dislocations move so as to reduce the thermal stress with a gradient in the insert caused by a large difference in the CTEs between tungsten and ODS-FS. This process includes a type of high temperature creep under thermal stress, and will continue until the transformation ceases or the thermal stresses are sufficiently reduced. A longer duration period for the phase transformation will promote creep and lead to a sufficient relaxation of the thermal stresses.

In this research, we investigated the effect of Phase transformation-induced creep (PTC) of insert material on the bonding strength of the dissimilar tungsten/ODS-FS joint.
2. Experimental Procedure

2.1 Materials for dissimilar joint

The materials used in this study were commercially available pure tungsten and an ODS-FS as the parent materials of joint, and three sorts of insert materials, such as S50C steel, pure iron and a V alloy, as listed in Table 1. The pure tungsten, which was called as S-TUN, was fabricated via HIP (Hot Isostatic Pressing) by Nippon Tungsten Co., Ltd., and exhibited an equiaxed grain structure. In the previous research, we compared the bonding fracture strength of several tungsten joints produced with commercially available different tungsten plates with deferent grain structures, and found that S-TUN showed the highest fracture strength among them. The ODS-FS was fabricated by mechanical alloying (MA) and hot extrusion.

2.2 Insert material

The S50C steel, containing 0.5 mass% C, undergoes the $\gamma \rightarrow \alpha$ phase transformation in the temperature range of 740–680°C as predicted by the equilibrium phase diagram of the Fe-C system. During cooling after the heat treatment of diffusion bonding, a large difference in the CTE results in the increase in thermal stress with decreasing temperature. On the other hands, the pure iron, containing 0 mass% C, undergoes the allotropic $\gamma \rightarrow \alpha$ phase transformation in the temperature of 911°C. It means that the pure iron insert has not coexistence region of two phase in the phase diagram.

2.3 Diffusion bonding and heat treatments after bonding

The specimens for bending tests were machined by wire electro-discharge methods. The specimen dimensions for bending test were 10 mm × 10 mm × 2.0 mm for the tungsten and ODS-FS, and 10 mm × 10 mm × 1.0 mm for the insert materials. The specimen surfaces were mechanically polished with abrasive sanding papers of #500 to #4000 and buff-polished with a diamond paste with particle size of 1.0 μm.

The insert material was set between the tungsten and ODS-FS specimens for solid phase diffusion bonding, as shown in Fig. 1 (a). The bonding was conducted at 1240°C for 1 hr in a vacuum ($8.0 \times 10^{-3}$ Pa) with a hot press by applying stress 2% of plastic strain to the insert material for uniform contact of the parent materials with an insert material.

The tungsten/ODS-FS joint was heat-treated in four cooling patterns (CP) in the temperature range of the phase transformation followed by cooling down to ambient temperature, as shown in Fig. 2 and Table 2. The cooling patterns of CP-1 and CP-2 involve 10 cycles of heating and cooling between 800 and 600°C and the temperature patterns are typical for carbon steels to exhibit transformation super plasticity. These two heat treatments differ in heating and cooling rates; 650°C/min for CP-1 and 40°C/min for CP-2. On the other hand, CP-3 and CP-4 are not cyclic but monotonic cooling patterns. In the CP-3 pattern the temperature was maintained at 600°C for two hours after cooling from the bonding temperature with the same cooling rate as in the CP-2 pattern. In the CP-4 pattern the joint was slowly cooled at a rate of approximately 1.6°C/min from 800°C to 600°C.

2.4 Evaluation of mechanical properties

In order to assess the effect of the post-heat treatment on fracture strength at room temperature, the bonding strength of the diffusion bonded specimens, which measures 5.0 mm (long) × 2.4 mm (wide) × 0.5 mm (thick) and shown in Fig. 1(b), were evaluated by miniaturized four-point bending test method with a specially designed fixture of the upper and lower spans of 0.8 and 3.0 mm, respectively. The details of specimen setting is shown in Fig. 3, where the joint specimen

![Fig. 1 Tungsten/ODS-FS joint: (a) Materials setting up for diffusion bonding. (b) Geometry of specimen for bending test.](image)

![Fig. 2 Different cooling patterns (CP) of post diffusion bonding heat treatments.](image)

### Table 1  The materials used in this research.

<table>
<thead>
<tr>
<th>Material</th>
<th>Designation</th>
<th>Compositions (mass%)</th>
<th>Shape</th>
<th>Geometries (mm)</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tungsten</td>
<td>W-TiC</td>
<td>W-1.1%TiC</td>
<td>Round bar</td>
<td>1.5</td>
<td>HIQed Equiaxed grains</td>
</tr>
<tr>
<td></td>
<td>S-TUN</td>
<td>W, Purity: 99.95%</td>
<td>Rectangular block</td>
<td>25</td>
<td></td>
</tr>
<tr>
<td>Insert material</td>
<td>S50C steel</td>
<td>Fe-0.5%C-0.25%Si-0.75%Mn</td>
<td>Round bar</td>
<td>φ = 50</td>
<td>Hot worked product</td>
</tr>
<tr>
<td></td>
<td>Pure iron</td>
<td>Fe, Purity: 99.99%</td>
<td>Plate</td>
<td>1.0</td>
<td>Rolled</td>
</tr>
<tr>
<td></td>
<td>V alloy</td>
<td>V-1.4Y-7W-9Mo-0.7TiC</td>
<td>Plate</td>
<td>11 × 26 × 36</td>
<td>Rolled</td>
</tr>
<tr>
<td>Ferritic steel</td>
<td>15Cr-ODS-FS</td>
<td>Fe-15%Cr-2%W-0.2%Ti-0.35%Y2O3</td>
<td>Rod</td>
<td>φ = 20</td>
<td>MA-Swaged</td>
</tr>
</tbody>
</table>
was placed on the span edges so that the tungsten/ODS interface was allocated just center of the upper span rods. The flexural rate was $1 \times 10^{-3}$ mm s$^{-1}$.

Since it is expected that the heating during bonding at 1240°C for 1 hr may change the yield strength at room temperature of the insert materials, tensile tests were also carried out for the joints of which the gage section measures 5 mm length, 1.2 mm width and 0.5 mm thickness using a tensile test machine equipped with a shear-type load cell (capacity: 5 KN) and LVDT (Linear Variable Differential Transformer) for measuring displacement with the maximum displacement of ±2.5 mm. The initial strain rate was $1 \times 10^{-3}$ mm s$^{-1}$.

### 3. Results

Four point bending stress-deflection behavior of tungsten/ODS-FS joints with an S50C insert is shown in Fig. 4, where the effect of cooling pattern on the bending stress can be seen. All the joints broke in the elastic deformation region and the numbers shown in the figure are the average and maximum values of the bending fracture stress. The average values are taken from the results of several specimens of which the number is shown in the blanket. It is considered that the bending fracture stress corresponds to the bonding strength of the joint, because fracture initiates at a location of the bottom surface of the bend specimen close to the interface between tungsten and inter-diffusion area of tungsten without exhibiting ductile behavior, as shown in Fig. 5. The average strengths of the specimens for CP-1 and CP-2 were 260 and 364 MPa, respectively, which shows an increase by 40% with decreasing cooling rate. On the other hand, the average strength for CP-3 of a monotonic cooling pattern with the same rate as CP-2 was 287 MPa, which was 21% lower than that of CP-2. It should be emphasized that a monotonic slow cooling pattern of CP-4 from 800 to 600°C yielded the highest strength among the four post heat treatment patterns, 377 MPa on average and 478 MPa at the maximum. CP-4 provides a cooling rate of approximately 1/25 of CP-3. The point is the duration time in the phase transformation temperature range, and CP-4

### Table 2. Details of cooling patterns of post diffusion bonding heat treatments.

<table>
<thead>
<tr>
<th>Tungsten Insert</th>
<th>Cooling Pattern after bonding</th>
<th>Temperature (°C)</th>
<th>Temperature history</th>
<th>Cooling/heating rate</th>
</tr>
</thead>
<tbody>
<tr>
<td>W-TiC Pure Fe</td>
<td>non</td>
<td>CP-1</td>
<td>800→600</td>
<td>Cyclic change: 10.5 times</td>
</tr>
<tr>
<td></td>
<td></td>
<td>CP-2</td>
<td>800→600</td>
<td>Cyclic change: 10.5 times</td>
</tr>
<tr>
<td>S-TUN S50C</td>
<td>non</td>
<td>CP-3</td>
<td>800→600</td>
<td>Fast cooling and holding at 600°C</td>
</tr>
<tr>
<td></td>
<td></td>
<td>CP-4</td>
<td>800→600</td>
<td>Slow cooling</td>
</tr>
</tbody>
</table>

![Fig. 3 Miniature four-point bending test fixture (mm).](image3.png)

![Fig. 4 Four point bending stress-deflection curves of tungsten/ODS-FS joints with S50C insert. The average was taken from the results of several specimens of which the number is shown in the blanket.](image4.png)

![Fig. 5 Bend fractured area observed from the bottom surface of the tungsten (S-TUN)/ODS-FS joints with pure iron insert (a), with an elemental mapping by EPMA (b). Tungsten (S-TUN)/ODS-FS joints with S50C insert (c) after heat treatment of CP-2, with an elemental mapping by EPMA (d).](image5.png)
offers the longest duration. The ongoing transformation during cooling induces stress relaxation, which is ascribed to time-dependent creep driven by thermal stress in the insert, and hence the maximum stress relaxation can be achieved in the CP-4 pattern. In view of the effectiveness in thermal stress relaxation, cyclic patterns, which include heating across the transformation temperature range, may not be advantageous, since the driving force for the relaxation of the stresses due to CTE difference between tungsten and ODS-FS decreases as the $\alpha \rightarrow \gamma$ transformation proceeds with increasing temperature.

Figure 6 shows the strengths of equiaxed-axis-grained tungsten/ODS-FS joints using three insert metals which possess the required properties such as high melting point, low activation etc. The tungsten/ODS-FS joint with the V insert was broken during specimen cutting before bending tests: the joint exhibited an extremely low bonding strength. On the other hand, the joint with the pure iron insert was able to be cut into bending specimens: the bonding strength was 324 MPa on average and 384 MPa at the maximum. In comparison to the bonding with the insert of V alloy or pure iron, the bonding with S50C steel insert under the CP-4 pattern yielded significantly high bonding strength. The percentages in the parentheses right next to the maximum stresses show the ratio of the bonding strength to the bending fracture strength (670 MPa) of S-TUN, 57% for the pure iron and 71% for the S50C insert. The bonding strength has not reached the fracture strength of S-TUN. Thermal stress due to CTE difference between tungsten and ODS-FS increases with decreasing temperature and is maximized at room temperature.

4. Discussions

4.1 Characteristic of insert materials

Insert materials are required to have a rather low yield strength for plastic thermal stress relaxation. The yield stresses of V alloy, S50C and pure iron at room temperature after the identical heat treatment condition of bonding (1240°C, 1 hr) were measured in Fig. 7. The strength of pure iron is significantly low, only 64 MPa. The strengths of S50C and V alloy were 374 MPa, 580 MPa, respectively. In terms of yield strength at room temperature, pure iron may exhibit the greatest stress relaxation, followed by S50C steel and V alloy. Nevertheless, the bonding with S50C instead of pure iron resulted in the highest bonding strength of the joint. This supports the model that a significant thermal stress relaxation can be operative through the creep during the $\gamma \rightarrow \alpha$ phase transformation and this mechanism is dominantly effective in the reduction of the residual thermal stress of the tungsten/ODS-FS joint.

4.2 Evaluation of bonding strength

The effect of cooling pattern was obviously observed in the bonding strength measured by bending test at ambient temperature. Figure 8 shows the plots of bending strength versus the total duration time of the $\gamma \rightarrow \alpha$ phase transformation during cooling after the diffusion bonding. Here, the $\gamma \rightarrow \alpha$ transformation occurs in the temperature region from 770 to 720°C ($\Delta T = 50^\circ$C). There is a good correlation between the measured bending strength and the total duration of the $\gamma \rightarrow \alpha$ transformation, indicating that thermal stress relaxation was induced by the phase transformation as described, where the total duration is critical for the extent of creep deformation. For an enough level of stress relaxation in the joint, a cyclic pattern that is known as a process using super plasticity does not seem prerequisite. In the general super plasticity experiment, tensile test is carried out under con-
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4.3 Thermal stress relaxation using phase transformation-induced creep

The thermal stress relaxation using phase transformation-induced creep (PTC) is discussed for the S50C insert material as follows: Fig. 9(a) illustrates the model of thermal stress reduction by $\gamma \rightarrow \alpha$ phase transformation induced creep in the S50C insert. During cooling after diffusion bonding, the large difference in the CTE of dissimilar materials results in an increase in thermal stress with decreasing temperature. The S50C steel undergoes the $\gamma \rightarrow \alpha$ phase transformation in the range of 740–680°C during cooling as shown in Fig. 9(b).¹⁵ The phase transformation generates dislocations in the $\alpha$ phase due to difference in specific volume between the $\gamma$ and $\alpha$ phases ($V_\gamma > V_\alpha$). The dislocations move so as to reduce the thermal stress in the stress gradient insert. In this process, the amount of plastic deformation increases with increasing thermal stress and increasing the duration spent for $\gamma \rightarrow \alpha$ phase transformation which offer a high temperature creep under thermal stress and will continue until the transformation ceases or the thermal stress is sufficiently reduced. A longer duration period for the $\gamma \rightarrow \alpha$ phase transformation may increase the amount of creep and lead to a sufficient relaxation of the thermal stress. Thus, the total duration is critical to develop creep. For an enough level of stress relaxation in the joint, a cyclic pattern does not seem to be prerequisite, instead simple cooling with a very slow rate in the dual phase region is more effective and considered to be adequate as the post heat treatment after bonding.

The obtained results in this research indicate that the thermal stress relaxation using insert materials that exhibit $\gamma \rightarrow \alpha$ phase transformation-induced creep results in a high bonding strength with increasing in the total duration time of the $\gamma \rightarrow \alpha$ transformation during cooling from the bonding temperature. Although the $\gamma \rightarrow \alpha$ transformation occurs both in pure iron and S50C, Fig. 8 shows a clear difference between the two in effectiveness in the reduction of thermal stress. The characteristic feature in the thermal stress relaxation which utilizes $\gamma \rightarrow \alpha$ phase transformation-induced creep is a coexistence of two phases of $\gamma$ and $\alpha$. S50C steel has two phases in the temperature range from 740 to 680°C, whereas pure iron exhibits the allotropic transformation point (910°C). In addition, the pure iron insert experiences a wider single phase temperature region from 910°C to room temperature, which may cause a greater buildup of thermal stress. Thus, the stress relaxation with pure iron insert is considered less than the relaxation with S50C insert. The advantage of low carbon steel as an insert is that a strict cooling condition setting is not necessary for the dual phase region. As for the effect of cyclic pattern of temperature history, which include heating operations across the transformation temperature range, it may not be advantageous for carbon steels, since the $\alpha \rightarrow \gamma$ transformation appears to increase thermal stress. Finally, it is expected that low and medium carbon steels (0.1–0.6%C) would be adequate as an insert for tungsten/ODS-FS joint with taking account of the transformation temperature range from A3 to A1 in order to gain a realistic and effective stress relaxation by $\gamma \rightarrow \alpha$ phase transformation-induced creep.

5. Conclusions

A cooling process with effective thermal relaxation was developed for bonding of tungsten and ODS-FS after diffusion bonding at 1200°C. The bonding strength of the tungsten/ODS-FS was assessed by 4-point bending tests. The main results obtained are as follows:

1) The bonding strength of the S-TUN/S50C/ODS-FS depends on the cooling pattern across the $\gamma \rightarrow \alpha$ transformation temperature range. A monotonic slow cooling at 1.6°C/min leads to the highest bonding strength of 478 MPa.

2) There is a good correlation between the measured bonding strength and the total duration of the $\gamma \rightarrow \alpha$ transformation during cooling. It is proposed that the stress relaxation is due to the $\gamma \rightarrow \alpha$ transformation induced creep under thermal stress during cooling. The $\gamma \rightarrow \alpha$ transformation induced creep will continue until the transformation ceases or the thermal stress is sufficiently reduced.

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1218–1222.