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Solid State Diffusion Bonding of Doped Tungsten Alloys with Different Thermo-Mechanical Properties

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To develop the joints using W materials with different thermo-mechanical properties, solid state diffusion bonding between two different W materials (pure W, K-doped W, and K-doped W-3%Re) with pure V interlayer (1.5 mm, 0.5 mm, and 0.05 mm) was carried out at 1250 °C. Thin interlayer was clarified to be desirable from the view point of the strength and thermal diffusivity. Diffusion bonding at lower temperature or utilizing the W materials with higher recrystallization temperature were also clarified to be desirable because pure W could recrystallize at 1250 °C. Therefore, it is pointed out that further evaluation based on the present work under the wide range of interlayer thickness and thermo-mechanical test conditions are necessary to obtain the optimum W/V/W joints.

Keywords: Solid state diffusion bonding, Tungsten, Potassium doping, Rhenium addition, Thermal diffusivity, Tensile strength

1. Introduction

Tungsten (W) is a primary candidate material for fusion reactor divertor because of its high melting point, thermal conductivity, sputtering resistance, and low tritium inventory [1]. In the case of the ITER monoblock divertor targets using pure W, the steep temperature gradient from heat-loaded surface to coolant of the divertor target and its periodical variation due to cyclic heat loading could induce complicated degradation behavior. Crack formation caused by thermal fatigue, thermal shock, and recrystallization embrittlement (see fig. 1 (a) [2]) and large deformation caused by plastic strain accumulation and high temperature creep [3] could occur at region near the heat-loaded surface. In contrast, brittle fracture at region near the coolant is one of the supposed issues because W materials show low temperature embrittlement. Therefore, it is required to improve and optimize the thermo-mechanical properties of the divertor materials.

Development of flat-tile-type divertor targets (see fig. 1 (c)), which consists of two different materials (one material at the heat-loaded surface side and the other one at the coolant side) with and without an interlayer between them, is in progress to overcome the different material issues in those regions of the divertor targets [4, 5]. In the present work, we focused on the development of flat-tile-type divertor targets using two W materials with different thermo-mechanical properties. Recently, various hot-rolled W materials strengthened by potassium (K) doping for dispersion strengthening and rhenium (Re) addition for solid solution strengthening have been developed in Japan, which were pure W, K-doped W, W-1%Re, W-3%Re, and K-doped W-3%Re [6, 7]. The advantages of K-doping and Re-addition include suppression of recrystallization and improvement of mechanical properties; however, addition of Re caused a decrease in the thermal conductivity. To fabricate the joints using two different W materials and to suppress any reaction layers and large defects, solid state diffusion bonding technique was applied in the present work. This paper describes fundamental thermo-mechanical properties of the joints using two different W materials and discusses further optimization of joints.

2. Experimental procedure

Rolled plates with 7 mm thickness of pure W, K-doped W, and K-doped W-3%Re were used in the present work, which were provided by A.L.M.T. Corp., Japan. These materials were fabricated by cold isostatic pressing, sintering, hot rolling, and final heat treatment at 900 °C for 20 min for stress relief.

Because it will be difficult to realize direct bonding between W materials below recrystallization temperature (around 1300 °C [8]), pure V sheets were employed as an interlayer between W materials. Thickness of V sheet were 1.5 mm, 0.5 mm, and 0.05 mm. The advantages of utilizing V are 1) complete solid solution in the W and V binary system and 2) low induced radioactivity of V. The V sheets were heat-treated at 400 °C for 1 h for dehydrogenation before bonding tests.

Figure 2 shows the schematic illustration and dimension of W/V/W joint. The W plate materials and V sheets were machined to a dimension of 20 mm^w × 20 mm^l × 5 mm^t and 25 mm^w × 23 mm^l × 1.5, 0.5, and 0.05 mm^t, respectively The nomenclature of the W material direction, L.D., T.D., and S.D., are as shown in fig. 2. The L.D. and S.D. correspond to the rolling direction (R.D.) and the direction along the thickness of the plate.

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In the diffusion bonding tests, the L-T surfaces were bonded under the mechanical press along the S direction. The combinations of W materials (materials A and B shown in fig. 2) and V interlayer thickness of the joints (joint #1 – #4) are summarized in table 1.

The solid state diffusion bonding was carried out in vacuum using a uniaxial hot-press testing machine (model Hi multi of Fuji Dempa Kogyo Co., Ltd., Japan) of national institute for fusion science (NIFS) in Japan. Test temperature, pressure, and holding time were 1250 °C, 20 MPa, and 1 h, respectively. These test conditions are similar to those of the previous research by Noto et al. [9], where joint between pure W and pure V showed smooth interface without any reaction layers and large defects. The joints are wrapped with tantalum foil as an oxygen getter to suppress any oxygen effects during the bonding tests.

To evaluate the fundamental properties of joints, cross-sectional metallographic observation by an optical microscope, element analysis by an energy dispersive X-ray spectroscopy (EDS), tensile tests at R.T., 300 °C, and 700 °C in vacuum using small specimen (gauge section: 0.92 mm^{w} × 0.81 mm^{l} × 1.00 mm^{t}), and measurement of thermal diffusivity using rectangular specimen (5 mm^{w} × 5 mm^{l} × 1 mm^{t}) were carried out. At the temperature of R.T. and 300 °C, W and V are considered to show brittle and ductile fracture, respectively. In contrast, at the temperature of 700 °C, both W and V are considered to show ductile fracture. In addition, to evaluate grain growth behavior of each W material during heat treatment, grain size measurement at the L-S surface was conducted before and after the heat treatment at 1100 °C, 1300 °C, 1500 °C, 1800 °C, 2000 °C, and 2300 °C for 1 h.

3. Results and discussion

3.1 Grain growth of W materials by heat treatment

Heat treatment temperature dependence of the grain size measured at the L-S surface of pure W, K-doped W, and K-doped W-3%Re are summarized in fig. 3. The initial grain sizes of as-received materials were 110 × 20 µm in pure W, 40 × 10 µm in K-doped W, and 28 × 8 µm in K-doped W-3%Re, which were plotted at 900 °C (heat treatment temperature of as-received W materials for stress relief) in fig. 3. Pure W showed significant grain growth above 1000 °C and its saturation above 1500 °C, which are attributed to the recrystallization. The grain size after recrystallization was 110 – 120 µm. In contrast, K-doped W showed almost no change of grain size up to 1300 °C and relatively small grain growth above that. The grain size after recrystallization was 30–35 µm. In the case of K-doped W-3%Re, monotonic increase of grain size above 1500 °C and no saturation of grain growth up to 2300 °C were observed.

Based on these evaluations, the pure W could be recrystallized during the bonding tests. In contrast, K-doped W and K-doped W-3%Re could show almost no grain growth and no recrystallization.

3.2 Cross-sectional observation and element analysis

Cross-sectional macroscopic images of the joints #1, #2, and #4 by optical microscope are shown in fig. 4 (a – c). All the joints (including the joint #3) showed smooth interface without any reaction layers and large defects. The EDS surface analysis of the joint #4 shown in fig. 4 (d-1 – d-3) exhibited absence of inter-metallic compound, which was attributed to the complete solid solution in the

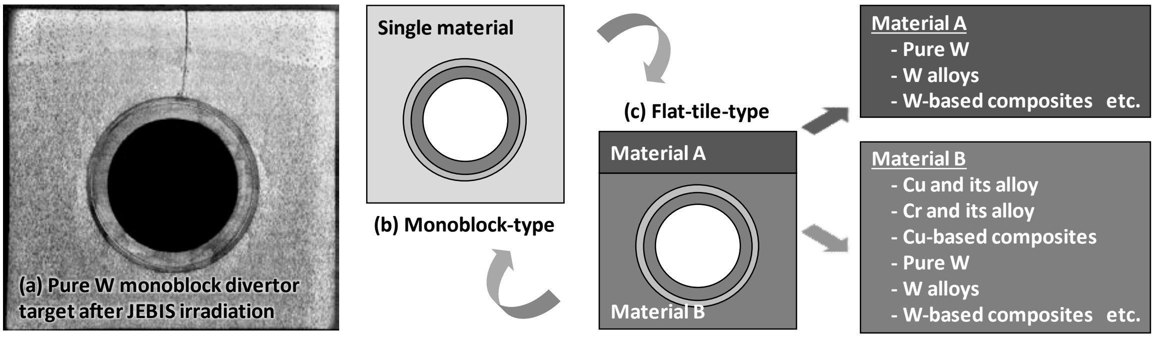


Fig. 1 (a) Cross-sectional metallographic image of monoblock-type divertor target using pure W after cyclic electron beam irradiation (20 MW/m^{2} x 300 cycles) using JEBIS [2], schematic illustrations of (b) monoblock-type divertor target using a single material and (c) flat-tile-type divertor target using various materials

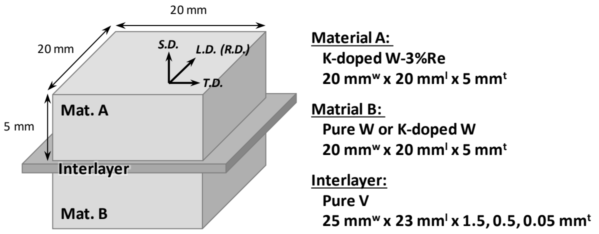


Fig. 2 Schematic illustration and dimension of W/V/W joint for solid state diffusion bonding

Table 1 Material combinations of W materials and V interlayer thickness of W/V/W joints

	Material A	Interlayer	Material B
Joint #1	K-doped W-3%Re	Pure V (1.5 mm^{t})	Pure W
Joint #2	K-doped W-3%Re	Pure V (0.5 mm^{t})	Pure W
Joint #3	K-doped W-3%Re	Pure V (0.5 mm^{t})	K-doped W
Joint #4	K-doped W-3%Re	Pure V (0.05 mm^{t})	Pure W

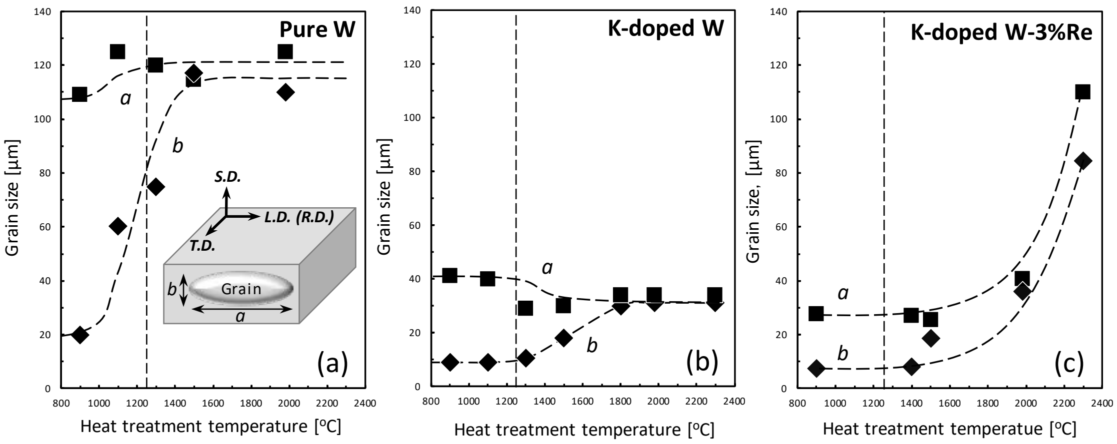
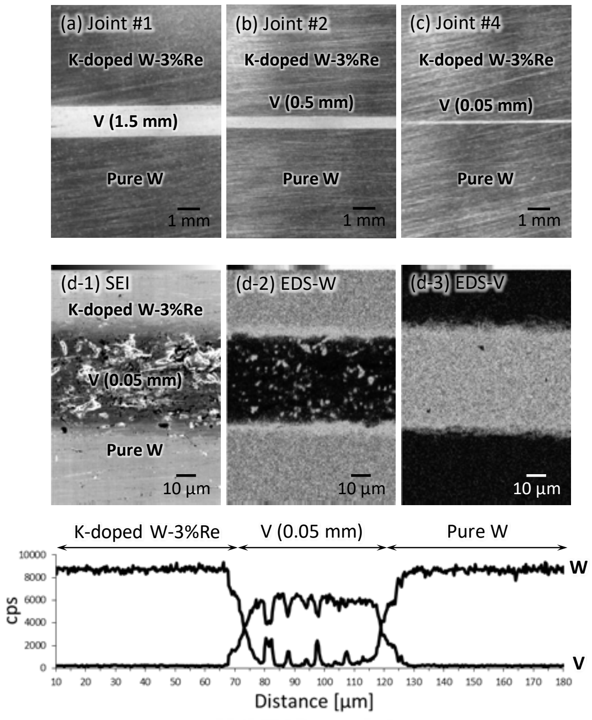


Fig. 3 Heat treatment temperature dependence of grain size (*a* and *b* shown as illustration in (a)) measured at L-S surface of (a) pure W, (b) K-doped W, and (c) K-doped W-3%Re



(d-4) EDS line-analysis

Fig. 4 Cross-sectional macroscopic images of (a) joint #1, (b) joint #2, and (c) joint #4 and (d) cross-sectional EDS analysis results of joint #4 ((d-1) secondary electron image, (d-2) W (Kα) map, (d-3) V (Mα) map, and (d-4) line-analysis result of W (Kα) and V (Mα))

W and V binary system. As shown in fig. 4 (d-4), the mutual diffusion length between W and V was approximately 15 μm.

3.3 Tensile properties

Fig. 5 (b) shows the ultimate tensile strength of the joints #2 at 700 °C, joint #3 at 700 °C, and joint #4 at R.T., 300 °C, and 700 °C. Fig. 5 (c – g) show the SEM images of ruptured tensile specimens. All the tensile specimens machined from the W/V/W joints showed no fracture at the bonded interfaces between W and V.

In the cases of the joints #2 and #3 at 700 °C, ductile fracture with necking occurred at the V interlayer region as shown in fig. 5 (f) and (g). The ultimate tensile strength of these joints was 150 – 160 MPa regardless of the difference of the W material (K-doped W-3%Re/V/pure W and K-doped W-3%Re/V/K-doped W). Therefore, the strength of joints #2 and #3 at 700 °C could be dominated by that of pure V (150 – 160 MPa) and that of pure W, K-doped W, K-doped W-3%Re, and bonded interfaces between W and V could be above 160 MPa at 700 °C.

In the case of the joint #4 at 700 °C, brittle fracture with no necking occurred at the pure W region as shown in fig. 5 (e). Fracture manner of the pure W was intergranular (shown in fig. 5 (e-3)). The ultimate tensile strength of this joint reached about 330 MPa although the strength of pure V could be 150 – 160 MPa at 700 °C based on the test results of the joints #2 and #3. According to the previous research on the weld joint of steel [10], strength of joint, whose weld metal has lower strength than that of base metal, increases with decrease in the weld metal thickness. For example, if the weld metal thickness is 40% of the thickness of tensile specimen, the strength of joint is 40% – 80% higher than that of weld metal. Because strength of pure V is lower than that of W materials and thickness of V interlayer of the joint #4 (0.05 mm) is only 5% of the thickness of tensile specimen (1 mm), it is possible that the strength of joint #4 (330 MPa) became much higher than that of pure V (150 – 160 MPa). Therefore, the strength of joint #4 at 700 °C could be dominated not only by that of W materials (about 330 MPa in pure W, above 330 MPa in K-doped W-3%Re), V interlayer (150 – 160 MPa), and bonded interfaces (above 330 MPa) but by thickness of the V interlayer.

In the case of the joint #4 at R.T. and 300 °C, brittle fracture with no necking also occurred at the pure W region as shown in fig. 5 (c) and (d). Fracture manner of the pure W was intergranular in both test temperatures (shown in fig. 5 (c-3) and (d-3)). The ultimate tensile strength of this joint was about 130 MPa at R.T. and 310 MPa at 300 °C. Based on the previous research by Fukuda et al. [6], where the same W materials as the present work were used, the ultimate tensile strength of pure W, K-doped W, and K-doped W-3%Re along the S direction showed monotonic decrease with decrease in test temperature below 500 °C. Therefore, the strength of joint #4 at R.T. and 300 °C could be dominated by the same mechanism as that at 700 °C and became smaller than that at 700 °C because of the reduction of the strength of pure W at lower temperatures.

3.4 Thermal diffusivity

Thermal diffusivity of the W/V/W joints was measured to evaluate the thermal property of the joints and existence of intrinsic defects inside of the joints, especially at the bonded interfaces. Four kinds of mono-layered specimens (pure W, K-doped W, K-doped W-3%Re, and pure V), two kinds of bi-layered specimens (pure W/V and K-doped W-3%Re/V), and three kinds of three-layered specimens (K-doped W-3%Re/V/Pure W, K-doped W-3%Re/V/Pure W, and K-doped W-3%Re/V/K-doped W) were machined from the joints #1 – #4. The thicknesses of individual layers are shown in the schematic illustration of fig. 6.

Vanadium volume fraction dependence of thermal diffusivity at R.T. of these layered specimens is summarized in fig. 6. The thermal diffusivity values at R.T. of pure W, K-doped W, K-doped W-3%Re, and pure V by open literature [7, 11] are also shown in this figure. Because the four mono-layered specimens showed almost the same values as those in the open literature, thermal diffusivity measurement of the specimen machined from the joints might include no technical issues. In general, the thermal diffusivity of the layered specimens became small with increase in V volume fraction because that of pure V was only 15 – 25% of that of W materials. The thermal diffusivity of layered specimens, which include higher volume fraction of K-doped W-3%Re, showed relatively low thermal diffusivity in detail when the V volume fraction is around 50% because the K-doped W-3%Re has approximately 35% lower thermal diffusivity than pure W and K-doped W. These tendencies correspond to the theoretical estimation. Thus, existence of intrinsic defects inside of the joints, which might degrade those thermal diffusivity, was expected to be negligible.

3.5 Discussion on optimization of W/V/W joint

Based on those evaluations mentioned above, the following results and issues are clarified.

1) The bonded interfaces between W and V were not the weakest at R.T., 300 °C, 700 °C regardless of the W materials and interlayer thickness.
2) The W/V/W joints using thick interlayer showed the fracture at pure V region at 700 °C.
3) The W/V/W joints using thin interlayer showed the fracture at W region at R.T., 300 °C, 700 °C.
4) The thermal diffusivity of the W/V and W/V/W layered specimens became small with increase in V volume fraction.

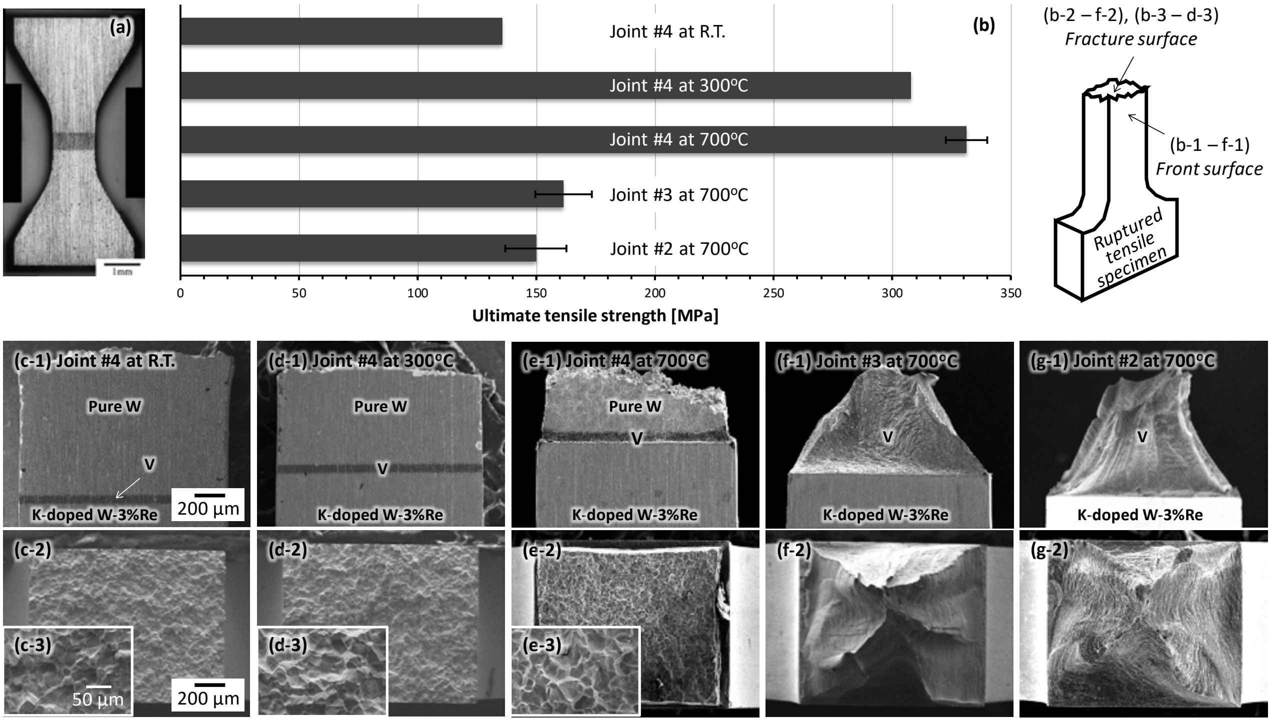


Fig. 5 (a) Tensile test specimen appearance machined from joint #1, (b) ultimate tensile strength of joint #2 at 700 °C, joint #3 at 700 °C, and joint #4 at R.T., 300 °C, and 700 °C and (c – g) SEM images of ruptured tensile specimens ((c-1 – g-1) front surfaces, (c-2 – g-2) and (c-3 – e-3) fracture surfaces, (c) joint #4 tested at R.T., (d) joint #4 tested at 300 °C, (e) joint #4 tested at 700 °C, (f) joint #3 tested at 700 °C, (g) joint #2 tested at 700 °C)

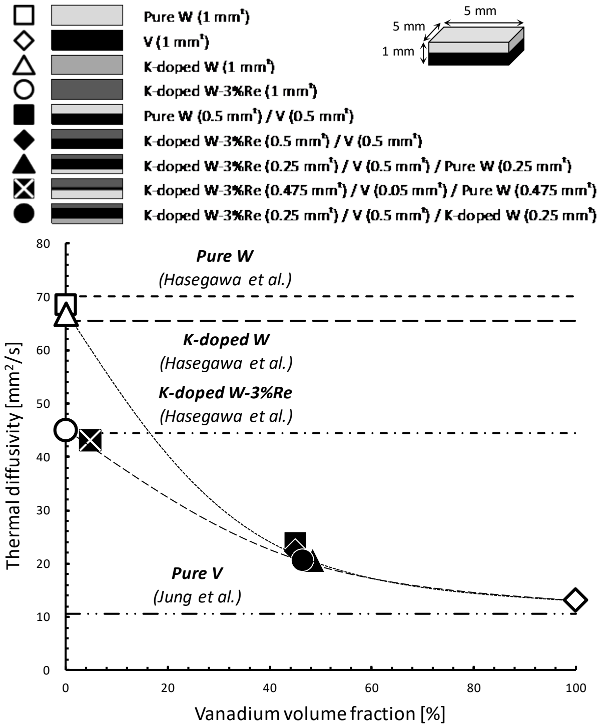


Fig. 6 Vanadium volume fraction dependence of thermal diffusivity at R.T. of W, V, W/V, and W/V/W layered specimens machined from various W/V/W joints

Because one of the most important functions of the divertor is heat conduction capability, degradation of the thermal properties in comparison with the parent materials (W materials in the present work) should be suppressed. In addition, it is desirable that the joint shows similar strength to the parent materials with no fracture at the bonded interface and interlayer. Therefore, it is better to reduce the V interlayer thickness in the case of W/V/W joints from the view point of the tensile properties and thermal diffusivity. Since the range of interlayer thickness and thermo-mechanical test conditions is limited in the present study, further evaluation is necessary for optimization.

From the view point of recrystallization of the W materials, reduction of the temperature of diffusion bonding (1250 °C in the present work) is desirable because the joint #4 showed the intergranular fracture of the pure W, which might be due to the recrystallization embrittlement. Reiser et al. [12] developed the W-foil laminate using V interlayer by the diffusion bonding at 900 °C for 1 h, which showed the better Charpy impact properties than that of the parent material (pure W). Thus, the diffusion bonding at 900 °C, where all the W materials could not show the recrystallization, can come in the sight. However, Liu et al. [13] pointed out that the diffusion bonding between W and V at 1050 °C for 1 h was not enough to realize the reasonable diffusion length because solid state diffusion bonding processes are usually conducted at temperature range of 0.5–0.8 T_m (T_m is melting temperature in K). Therefore, the optimum temperature from 1050 °C to 1250 °C and holding time of diffusion bonding should be found to realize the reasonable diffusion length with no recrystallization if the pure W is utilized for the W/V/W joints or the W materials with higher recrystallization temperature (K-doped W and K-doped W-3%Re etc.) should be utilized instead of the pure W.

4. Summary

To develop the joints using W materials with different thermo-mechanical properties for the flat-tile-type divertor targets, solid state diffusion bonding between different W materials (pure W, K-doped W, and K-doped W-3%Re) with pure V interlayer (1.5 mm, 0.5 mm, and 0.05 mm) was carried out at 1250 °C for 1 h. All the W/V/W joints showed smooth interface without any reaction layers and large defects. The mutual diffusion length between W and V was approximately 15 μm. The strength and thermal diffusivity of joints increased with decrease in the interlayer thickness. Thus, it is pointed out that the thin interlayer is desirable for the W/V/W joints. However, the pure W showed recrystallization after the diffusion bonding at 1250 °C. Therefore, it is also pointed out that the lower temperature of diffusion bonding and utilizing the W materials with higher recrystallization temperature should be considered for obtain the optimum W/V/W joints.

Acknowledgments

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Table 1 Material combinations of W materials and V interlayer thickness of W/V/W joints

	Material A	Interlayer	Material B
Joint #1	K-doped W-3%Re	Pure V (1.5 mm^t)	Pure W
Joint #2	K-doped W-3%Re	Pure V (0.5 mm^t)	Pure W
Joint #3	K-doped W-3%Re	Pure V (0.5 mm^t)	K-doped W
Joint #4	K-doped W-3%Re	Pure V (0.05 mm^t)	Pure W