# **Mechanism of Diffusion Bonding**

#### Subjects: Materials Science, Characterization & Testing

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Critical aspects of innovative design in engineering disciplines like infrastructure, transportation, and medical applications require the joining of dissimilar materials. Welding and brazing, while widely used, may pose challenges when joining materials with large differences in melting temperature and can lead to mechanical property degradation. In contrast, diffusion bonding offers a lower temperature process that relies on solid-state interactions to develop bond strength. The joining of tungsten and steel, especially for fusion reactors, presents a unique challenge due to the significant disparity in melting temperatures and the propensity to form brittle intermetallics. Here, diffusion characteristics of tungsten—steel interfaces are examined and the influence of bonding parameters on mechanical properties are investigated. Additionally, CALPHAD modeling is employed to explore joining parameters, thermal stability, and diffusion kinetics. The insights from this research can be extended to join numerous dissimilar materials for specific applications such as aerospace, automobile industry, power plants, etc., enabling advanced and robust design with high efficiency.

diffusion bonding

dissimilar materials joining CALPHAD

Tungsten steel

plasma facing materials

# **1. Introduction**

Technological advancement depends on the complex design of superior materials. The effectiveness of such design requires a site-specific material distribution and this often calls for a proper solution of joining two or more materials that is capable of providing the necessary mechanical and microstructural integrity of the design <sup>[1]</sup>. One such design is the structure of fusion reactors. Fusion reactors are potentially capable of meeting the need of the ever-increasing energy demand while keeping the environment pollution-free owing to its fuel abundance, zero carbon emission, and potential for economical operation <sup>[2][3]</sup>. However, several issues need to be addressed in order to make it feasible to generate energy from fusion reactors in a safe manner <sup>[4]</sup>. One of the major problems is the joining of plasma facing materials and structural materials of fusion reactors <sup>[5][6][7][8]</sup> Among the plasma facing materials and structural materials of fusion reactors <sup>[5][6][7][8]</sup>. However, being a brittle material materials (PFM), tungsten is considered to be the most promising candidate material. Some of tungsten's major advantageous mechanical properties include high melting temperature, high thermal conductivity, low vapor pressure, low tritium retention, high sputtering resistance, and so on <sup>[9][10][11]</sup>. However, being a brittle material at room temperature and with a high ductile brittle transition temperature, tungsten is not capable of providing structural integrity for the reactor <sup>[12][13]</sup>. As a structural material, ferritic/martensitic (FM) steel is one of the best

candidate materials owing to its superior mechanical properties capable of providing structural integrity <sup>[14]</sup>. Therefore, it is necessary to find a suitable solution for joining tungsten and ferritic/martensitic steel.

Even though numerous studies with different designs and approaches have been explored to join tungsten with steel [15][16][17], an effective and real-life applicable method to join these two materials is yet to be found. Some major issues of joining tungsten with steel are the difference between melting temperatures (1550 °C for FM steel vs. 3400 °C for W) which prevents conventional welding, a large difference in coefficients of thermal expansion (CTE) ( $12 \times 10^{-6} \text{ K}^{-1}$  for FM steel vs.  $4.4 \times 10^{-6} \text{ K}^{-1}$  for W), and formation of brittle intermetallic phases like Fe<sub>7</sub>W<sub>6</sub> (µ-phase) [18][19][20][21]. A proper joining of tungsten and steel may require the introduction of multiple interlayers in between them which would help to lower the mechanical property gradient along the joining interfaces and prevent the formation of brittle intermetallics. CALPHAD (CALculation of PHAse Diagram) modelling, which plays a crucial role in understanding complex materials' systems under different conditions, has been leveraged for a better understanding of tungsten steel joining [22]. CALPHAD is a widely used computational approach in materials science to predict phase equilibria in multi-component systems [23]. It utilizes thermodynamic databases containing Gibbs energy data for phases as a function of temperature, pressure, and composition.

# 2. Mechanism of Diffusion Bonding

Understanding the atomic bonding formation through the interaction of atoms during diffusion is of paramount importance when investigating solid-state bonding in materials. The diffusion process is determined by the kinetics of atom migration through surface, grain boundary, or bulk matrix <sup>[24][25]</sup>. Manufacturing process parameters such as temperature <sup>[26]</sup>, pressure <sup>[27]</sup>, holding time <sup>[28]</sup>, surface roughness <sup>[29]</sup>, and activation layers play a key role in the formation of the bond <sup>[30][31][32]</sup>. While investigating the joining of different configurations of tungsten and steel, one must pay careful attention to the process parameters and diffusion kinetics to have a good understanding of why different parameters yield different microstructural and mechanical properties.

# 2.1. Diffusion Bonding Overview

The definition of diffusion bonding provided by Kazakov <sup>[33]</sup> has been adopted by the International Institute of Welding (IIW): "Diffusion bonding of materials in the solid state is a process for making a monolithic joint through the formation of bonds at atomic level, as a result of closure of the mating surfaces due to the local plastic deformation at elevated temperature which aids interdiffusion at the surface layers of the materials being joined". Diffusion bonding between two mating surfaces goes through three steps <sup>[34]</sup>. In the first step of diffusion bonding, asperities from one mating surface exert force on another mating surface, which results in localized plastic deformation at the bonding temperature. Brittle oxides on the surface are fractured and metal-to-metal contact is established. In the second step, the thermally activated migration of abond. In the final step, elimination of the majority of pores occurs and a homogenized bond is produced <sup>[35]</sup>. A reaction layer is produced in the interface, and process parameters dictate the characteristic of this reaction layer. A large difference between diffusivity of major alloying element from the mating surfaces can result in Kirkendall pores and degrade the mechanical properties of

the joint <sup>[36]</sup>. In a tungsten-steel transition layer configuration, the presence of chromium and aluminum often results in the formation of a passivation oxide layer <sup>[37][38]</sup>. Therefore, care should be taken while determining process parameters like temperature and pressure so that these oxide layers can be eliminated and contact between the mating surfaces is established.

### 2.2. Advantage of Diffusion Bonding over Welding

The near net shape process of diffusion bonding has been explored widely because of the flexibility to choose a combination of elemental compositions while avoiding the formation of brittle intermetallic phases at the interface <sup>[36]</sup>. It is also possible to suppress elemental segregation in diffusion bonding and maintain a relatively low residual stress in comparison to other non-solid-state welding processes <sup>[39][40][41][42]</sup>. Diffusion bonding stands out among various joining techniques due to its minimal temperature gradient in the joint in comparison to other methods. Elimination of a heat-affected zone (HAZ) and highly complex microstructure from non-equilibrium phase transformation and chemical segregation during melting is possible via solid-state diffusion bonding, which makes it a lucrative option to be employed for joining in different industrial sectors like aerospace <sup>[43][44][45][46][47]</sup>, fusion reactor concepts <sup>[3][48][49][50][51][52]</sup>, plate type heat exchangers and reactors <sup>[53][54][55][56][57][58][59][60]</sup>, and so on <sup>[24]</sup>.

### 2.3. Atomistic Model of Diffusion

The atomic diffusivity is given by the following Equation (1), which represents the increase in diffusivity with the increase in temperature <sup>[24][61]</sup>:

$$D = D_o \exp\left[\frac{-\Delta E_{act}}{\mathrm{RT}}\right] \tag{1}$$

Here, *D* is the diffusivity (m<sup>2</sup>/s), *Do* is the frequency factor (m<sup>2</sup>/s), *Eact* is the energy of vacancy formation and migration (J/mol), RR is gas constant, and TT is temperature in Kelvin. The thickness of the reaction layer (*x*) in a diffusion couple is estimated by the following Equation (2) <sup>[62]</sup>.

$$x = kt^n \tag{2}$$

where *k* is a rate constant, *t* represents time, and *n* is a kinetic exponent. This equation is based on the solution of Fick's second law. For interfacial diffusion, grain boundary diffusion, and volume diffusion, the value of *n* is 1, 0.5, and 0.25, respectively  $\frac{[63][64][65]}{10}$ . In other words, the different values of *n* come from different diffusion pathways.

## 2.4. Important Parameters for Diffusion Bonding

#### 2.4.1. Temperature

The most significant factors to influence the diffusion bonding are bonding temperature, bonding pressure, and holding time. As it can be seen in Equation (3), the diffusivity is a function of temperature and it is assumed to follow an Arrhenius-type relation which can be shown in the following equation where the rate constant *k* from Equation (2) is presented as a function of temperature  $\frac{661}{2}$ .

$$k = k_o \exp\left[\frac{-E_a}{\mathrm{RT}}\right] \tag{3}$$

where ko is the pre-exponential factor and Ea is activation energy of reaction layer growth. A rise in the temperature tends to increase the thickness of the reaction layer <sup>[26]</sup>. With the temperature rise, there is an increase in diffusivity and the diffusion coefficient can be doubled even for a moderate rise of 20 °C <sup>[61]</sup>.

#### 2.4.2. Pressure

A minimum amount of bonding pressure is required for proper diffusion bonding at a particular temperature. The applied pressure is responsible for the first step of diffusion bonding where force is exerted on asperities to produce deformation and eliminate oxide layers. Several studies pointed out that a rise in pressure to moderate values results in a higher bond strength <sup>[67][68][69]</sup>. A few studies discussed impulse pressure-assisted diffusion bonding where varying pressure is used during diffusion bonding instead of a constant pressure and it was claimed that the overall processing and bonding time was reduced through the use of impulse pressure-assisted diffusion bonding <sup>[27][70][71][72][73][74][75][76]</sup>.

#### 2.4.3. Holding Time

The holding time is another very important parameter for determining the strength of diffusion bonding as it can be understood from Equation (2). In previous studies <sup>[28][77][78][79][80]</sup>, an increase in holding time increased the strength of the bond. Time is required to fill up the pores in the mating surface in order to establish a sound bond. Sufficient pressure at high temperatures causes deformation and holding time needs to be controlled in order to have the desired mechanical properties in the bond. A long heating time often results in excessive loss of intermediate layers and, as a consequence, a significant deterioration of the dissimilar materials' joint, which produces a weaker bond. On the other hand, if the holding time is too short, then there may be insufficient solid-state diffusion, which would also cause a weaker bond. Models have been developed that can estimate the required time for proper diffusion bonding. For example, Li <sup>[81]</sup> developed a probabilistic model using Fick's diffusion law for the necessary time to ensure sufficient diffusion. By following Equation (4), total bonding time *t* can be estimated.

$$t = \int_{f_0}^{f} \frac{-\sigma_e/\dot{\epsilon}}{\left(\left(3p\sqrt{f} - \frac{2\gamma\sqrt{f}}{r_o} + \frac{6\gamma}{r_o}\right) + \frac{2r_o^2p}{\ln\left(\frac{1}{f}\right) - \frac{1-f}{2}} - \frac{\Omega}{KT} \left[\frac{1 + \frac{h}{d}}{h} D_{\rm gb}\delta + 2\Omega D_{\rm v}\right]\right)} df \tag{4}$$

Here,  $\Omega$  is the atomic volume; *D*gb is the grain boundary diffusivity; *D*v is the volume diffusivity; and  $\delta$ , *d*, and *h*, are grain boundary width, grain size, and void height, respectively. *r*O is the initial void radius or surface roughness, *r* is the instantaneous void radius,  $f = \left(\frac{r}{r_o}\right)^2$  is the surface energy,  $\sigma e$  is the Levy–Mises effective stress,  $\epsilon$ . is the effective plastic strain rate, *K* is the Boltzmann constant, *p* is the external pressure, and *T* is the temperature in Kelvin.

#### 2.4.4. Other Diffusion Parameters

Other important factors that influence the diffusion bonding are surface roughness and passivation layers. Larger asperities need to be removed in order to create a sound bond [56][57]. A good surface finish is desired because the initial contact area largely depends on the presence of asperities and voids on the mating surfaces [78]. However, a contradictory result was found by Zuhuri <sup>[29]</sup>, where it is shown that a rougher surface yielded a better tensile strength of the bond. As a possible explanation, it was mentioned that a rougher surface would have more asperities and, therefore, exerted pressure would be more localized to result in higher force that will eventually cause a higher diffusion between the mating surfaces. A surface roughness of 0.84 µm produced a maximum ultimate tensile strength (UTS) value of ~130 MPa, while a surface roughness of 0.18 µm produced a UTS value of ~80 MPa for the bonding of aluminum alloys. On the other hand, tensile strength and joint efficiency were improved with a smoother surface finish [82][83]. The tensile strength of the joint of a Cu-Cu (copper-copper) couple was found to be ~50 MPa and ~5 MPa for surface roughnesses of 90 nm and 1.5 µm, respectively, for a bonding temperature of 400 °C and 20 min bonding time. It was also suggested that a higher pressure is required for a superior bonding when the surface roughness is high [84]. The passivation layer becomes an issue for materials like aluminum, stainless steel, nickel, and titanium alloys where a thin oxide layer prevents further oxidation of the materials, resulting in good corrosion resistance <sup>[61]</sup>. For materials like titanium, it is possible to dissolve the passivation layer in the matrix material [46]. For other materials, processes like chemical pickling or sputtering with argon ions can be used to remove passivation lavers [61].

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