Diffusion Bonding: Influence of Process Parameters and Material Microstructure

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Additional information is available at the end of the chapter

Abstract

Diffusion welding is a solid joining technique allowing for full cross-section welding. There is no heat-affected zone, but the whole part is subjected to a heat treatment. By diffusion of atoms across the bonding planes, a monolithic compound is generated.

The process takes place in a vacuum or inert gas atmosphere at about 80% of the melting temperature and is run batch-wisely. Hence, it is rarely used despite its advantages to achieve holohedral joints and is widespread in the aerospace sector only.

The quality of a diffusion-welded joint is determined by the three main parameters bonding temperature, time, and bearing pressure. The difficulty tailoring the process is that they are interconnected in a strong nonlinear way.

Several additional factors may influence the result or may change the material, e.g. surface roughness and passivation layers, all kinds of lattice defects, polymorphic behaviour, and formation of precipitations at grain boundaries, design of the parts to be welded and its aspect ratio as well as mechanical issues of the welding equipment. Hence, experiments are necessary for almost each special part.

In this chapter, an overview about the experience of diffusion welding is given. Influences are discussed in detail and conclusions are derived.

Keywords: diffusion welding, diffusion bonding, lattice defects, grain growth, precipitation, sensitization, passivation layer

1. Introduction

Diffusion welding is the only welding technique by means of which full cross-sectional welds, also of internal structures, can be obtained. Normally, there is no liquid phase and the monolithic compound is formed completely under solid-state conditions.

For the conditions to be appropriate, mechanical properties across the joined part are comparable to the bulk material. Due to heating of the whole parts, no distinct heat-affected zone (HAZ) is formed. However, properties are changed compared to the as-delivered conditions of the material. This may cause problems in some cases.

For diffusion welding, special and expensive equipment is required: the parts have to be mated at high temperatures by applying high forces depending on the size and cross-section to be welded under a vacuum or inert gas environment. Equipment and parts are heated mostly indirectly by radiation. To limit thermal stress, the heating rates are restricted to some 10 K/min.

The welding process takes place in vacuum and cannot be performed on site. Mating surfaces must be free of any impurities and have a low surface roughness without deep scratches. Joining of multiple layers is possible in one step.

Diffusion welding is always accompanied by a certain deformation of the parts. This deformation depends mainly on bonding temperature, bonding time and bearing pressure. Unfortunately, influences of temperature and bearing pressure are non-linear, making it difficult to predict the deformation of a new design. Additionally, secondary impacts on deformation and the quality of joining may be due to specific geometric parameters, e.g., the aspect ratio, the number of layers, the micro-structure of the material itself and surface layers.

Recently, thin coatings of other metals, forming a temporary liquid phase (TLP) by passing a eutectic composition, or multiple layers of different metals of nanometre thickness exploiting the enormous interfacial energy of such compounds were investigated.

In contrast to conventional welding techniques, such processes are highly complex. The process has to be optimised for each material and even for different compositions of alloys depending on the geometry. For this reason, application of diffusion welding is limited to the aerospace industry or special applications where other welding techniques fail. For example, large- and thin-walled titanium sheets are joined to reinforcing structures and internal cooling channels for injection moulding tools and nozzles of rocket engines.

Unfortunately, not all the information necessary for reproducing the results, e.g., material, procedure of sample preparation and process parameters, is given in the literature.

For joining micro-structured components, additional aspects must be taken into account.

The aim of this chapter is to summarise knowledge on diffusion welding in conjunction with the fundamental processes taking place inside of the micro-structure of a material. For this, lattice defects are discussed according to their dimensionality.

2. Micro-structure of metals and the impact of lattice defects on diffusion welding

2.1. Atoms in the lattice of metals

To minimise the energy of a system, isolated metal atoms tend to arrange in a regular lattice at positions according to the annihilation of attractive and repulsive forces (**Figure 1**). The positions are well-defined and specific of each metal. Hence, they can be used, e.g., for determining the composition of an alloy by means of WDX (wavelength dispersive X-ray). When forming a compound, atoms split up into positively charged atomic nuclei, while valence electrons are transferred to the so-called electron gas and can move freely within the lattice. Consequently, metals are good conductors of heat and electricity.
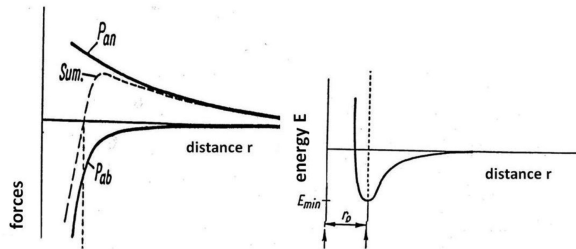
Figure 1. Equilibrium of attractive and repulsive forces in the metallic lattice [1].

2.1.1. Thermal expansion

Depending on the thermal energy of the whole system, the positively charged atomic nuclei oscillate around their position, leading to a thermal expansion (**Figure 2**). According to *Grüneisen's rule*, linear expansion is in the range of 2% and volumetric expansion is 6–7% up to the melting point of a metal [1]. Hence, the melting point can be used to estimate the thermal coefficient of expansion. Below the melting temperature, the oscillation amplitude is about 12% of the lattice constant [2].
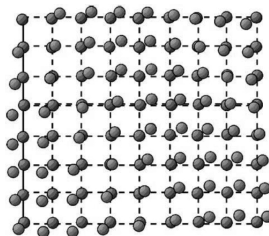

Figure 2. Thermal oscillation of atoms.

2.1.2. Thermal activation, diffusion, polymorphism and zero-dimensional lattice defects

With increasing thermal oscillation, not only the amplitude increases but also the energy of collisions between atoms. Gradually, some atoms are facilitated to leave its lattice sites and a vacancy is left leading to a punctual stress state (**Figure 3**). With increasing temperature, an exponentially increasing number of atoms is displaced from the lattice sites and the density of vacancies is considerably enhanced (Eq. (1)):

$$c_V = \frac{n}{N} * \exp.\left(\frac{-\Delta U}{RT}\right) \tag{1}$$
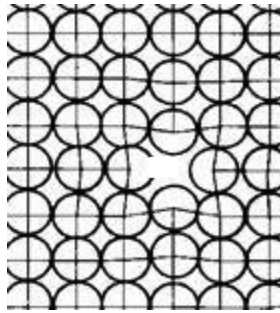
Figure 3. Vacancy in the lattice causing punctual stress.

where cV is the concentration of vacancies (cm^{-3}), n is the number of vacancies, N is the number of sites in the metallic lattice, U is the energy of formation of vacancies (for metals 80–200 J/mol), R is the gas constant (J/mol*K) and T is the temperature (K).

Vacancies are regular lattice sites not occupied by an atom. Due to a missing atom, the surrounding atoms tend to fill the gap and the lattice is distorted at this point, representing a zero-dimensional defect.

According to [3], the density of vacancies is 10^{-12} at room temperature and increases to 10^{-4} below the melting temperature.

Vacancies strongly facilitate the diffusion of atoms between different sites of the lattice and, hence, concentration facilitates the formation of a monolithic compound during diffusion welding. As a consequence, the coefficient of diffusion increases exponentially with temperature (Eq. (2)). An increase in bonding temperature by 20 K may result in a doubling of the diffusion coefficient, thus illustrating the strong non-linear influence of temperature on diffusion welding:

$$D = D_0 * \exp.\left(\frac{-\Delta U}{RT}\right) \tag{2}$$

where D is the diffusion coefficient (m²/s), D_0 is the frequency factor (material constant) (m²/s) and U is the energy of formation of vacancies (J/mol).

The number of vacancies versus temperature can be plotted as a logarithmic function, the so-called Arrhenius plot (**Figure 4**).
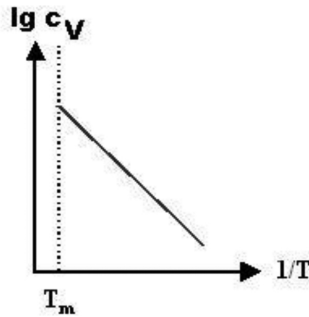

Figure 4. Arrhenius plot. The density of vacancies increases with a logarithmic dependency with temperature.

Depending on the real micro-structure of technical materials, different types of diffusion can be distinguished corresponding to different activation energies for different lattice defects. Straight lines for different diffusion paths can be plotted for surface, grain boundary and volume diffusion, respectively (**Figure 5**). For diffusion welding, grain boundary diffusion predominates at low and medium temperature. As the cross-section of grain boundaries is related to the volume and the density of vacancies increases exponentially, volume diffusion becomes predominant at high temperature.
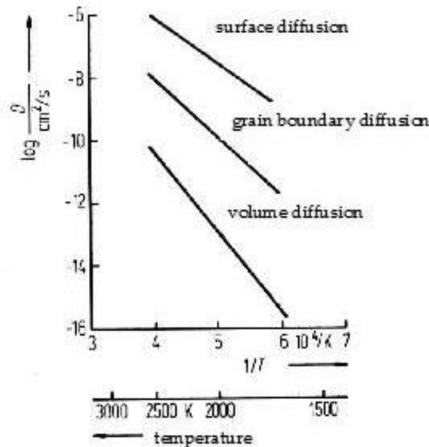

Figure 5. Different modes of diffusion of atoms versus temperature [1].

At the same time, grain growth takes place at high temperatures, which minimises the interfacial energy of the system. If the material shows no polymorphic transformation or the grain boundaries are not pinned by insoluble intra-granular precipitations (e.g., for ODS alloys), diffusion welding will be accompanied by grain growth.

Technical materials are no pure metals, but also contain other sorts of atoms, e.g., alloying elements like manganese, chromium or carbon for steel. Similar to vacancies, these atoms are integrated into the basic lattice as zero-dimensional defects. If they form the same type of lattice (e.g., cubic face-or cubic space-centred), and if the difference in atomic radii is less than 15%, they can occupy regular sites of the host lattice [4]. Small non-metallic atoms with an atomic radius smaller than 59% of the host atoms can be dissolved interstitially like carbon in iron [1].

Although solubility of interstitial atoms is low, they have diffusion coefficients higher by some orders of magnitude in the lattice, since more suitable gaps are available.

In case of low temperature rates during cooling down from diffusion welding temperature, this may be of relevance to the formation of undesired precipitations. The dwell time during diffusion welding should always be kept in the range of solution annealing for an alloy. This may conflict with a low temperature to limit grain growth.

If a metal is polymorphic, abrupt changes in solubility and in the diffusion coefficient may occur. For iron, e.g., these parameters change by two orders of magnitude (**Figure 6**). Reasons are different solubilities for foreign atoms and different sizes of gaps between the atoms in the lattice. For example, maximum solubility of carbon in α-ferrite (cubic space-centred) is 0.02% at 723°C, whereas the solubility of carbon in Y-ferrite (cubic face-centred) is 2.06% at 1143°C, i.e., higher by a factor of about 100 [1].
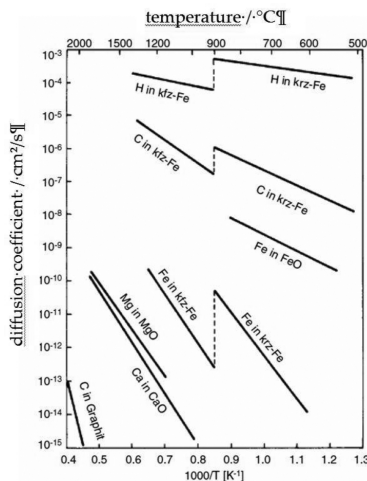

Figure 6. Diffusion coefficients of different sorts of atoms and depending on the type of lattice [5].

Additionally, polymorphism is accompanied by a complete new formation of the micro-structure, and grain size is reduced. Although grain growth occurs at high temperature, the same transformation happens when cooling down. This is the reason why normal steels, showing an a⇔Y transformation, can be diffusion welded easily with a finely grained micro-structure (**Figure 7**).
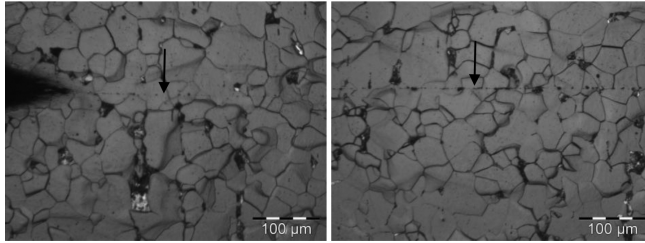
Figure 7. Diffusion weld of St 37 (1.0254), T = 1075°C, t = 1 h, p = 10 MPa, normal $\alpha \Leftrightarrow \gamma$-transformation, deformation: 3.13%.

On the other hand, a diffusion weld of austenitic steel is displayed in **Figure 8**. The impact of the four times longer dwell time on the grain size can be seen clearly.
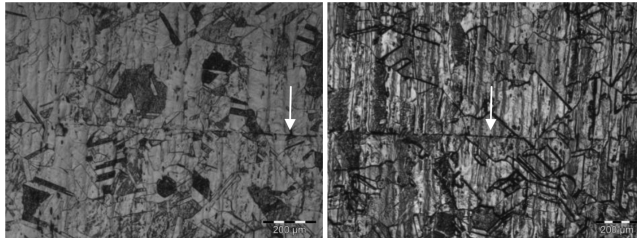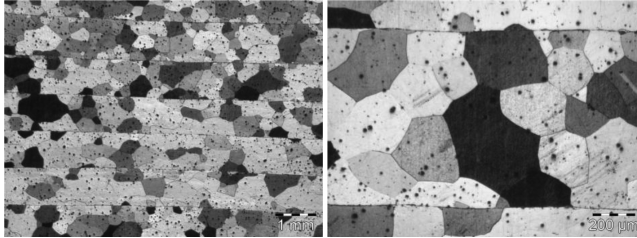

Figure 8. Diffusion weld of austenitic stainless steel AISI 304 (1.4301) at T = 1075°C, p ≈ 15 MPa. Left: t = 1 h, deforma-tion: 2.75%. Right: t = 4 h, deformation: 7.04%.

Although the bearing pressure for 1-h dwell time is 50% higher than for 1.0254, deformation is comparable due to the lower diffusion coefficient in the cubic face-centred lattice. When dwell time is increased by a factor of 4, however, deformation increases by a factor of about 2.5, see [6].

In **Figure 9**, ten 1-mm layers with a diameter of 40 mm of a fully ferritic stabilised stainless steel were diffusion welded. Welding at T = 1075°C, t = 1 h and p = 10 MPa for comparison failed due to excessive deformation. Even at a reduced temperature of T = 1000°C and reduced bearing pressure of p = 6 MPa [7], the deformation was huge at 14.6%. For T = 950°C, t = 1 h, p = 6 MPa, the deformation was still 3.8%.

These high deformations under these mild conditions have to be attributed to the high diffusion coefficient in ferrite, see **Figure 6**. However, despite the high deformation and

excessive grain growth due to lacking polymorphism, only very little grain growth across the bonding planes is visible in **Figure 9**, illustrating the role of surface passivation layers, see Section 2.2.

Figure 9. Diffusion weld of a fully ferritic stabilized stainless steel Crofer 22 APU (1.4760) at T = 1000°C, t = 1 h, p = 6 MPa. Deformation: 14.6%.

*2.1.3. One-dimensional defects: impact of dislocation density on mechanical properties*

Dislocations represent an inserted plane in a metallic lattice (**Figure 10**).
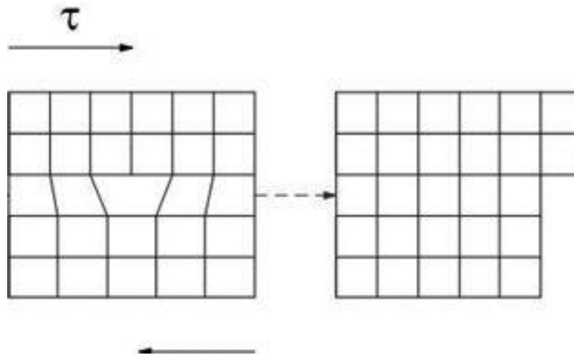
Figure 10. Plastic deformation by the movement of a dislocation across the lattice.

Of course, the inserted plane does not end at a constant level in different layers of the third dimension, but at an arbitrary depth, leading to complex stress conditions. If adequate shear stress appears, the dislocations are moved through the lattice in a step-wise manner, thus causing a plastic deformation. However, the dislocation density is not dropping, despite the dislocations leave the material at the surface. In opposite, it increases exponentially during cold working due to the so-called Frank-Read mechanism. Dislocation density can be given as a length of the dislocation line per unit of volume and can reach as much as 10^{12} cm^{-2} [2]. Only by cold work hardening alone can the mechanical strength of a material be multiplied (**Figure 11**).
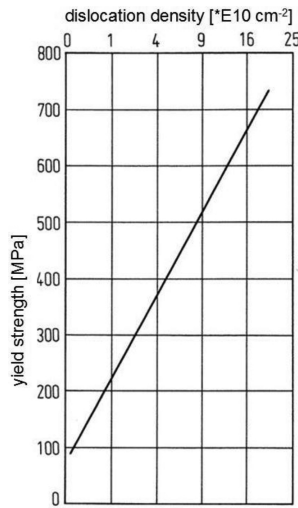
Figure 11. Increase of yield strength of pure iron by cold work hardening [8].

At room temperature, dislocation movement is the predominant deformation mechanism in metals. Dislocations represent a one-dimensional lattice defect.

Dislocations mean an energy excess compared to an undistorted lattice. Hence, at elevated temperatures of about 40% of the melting temperature of pure metals and 50% for alloys, recrystallization takes place [9]. For metals showing no polymorphism, cold work hardening and subsequent recrystallization is the only way to reduce the original grain size. However, it is applicable to half-finished products only.

Hence, when cold-worked material is diffusion welded, recrystallization will be included and affect the grain size.

2.1.4. Two-dimensional defects: grain and phase boundaries; interfacial layers and their influence on diffusion welding

Two-dimensional defects of a metallic lattice are reflected, e.g., by grain boundaries. They can be described as an interfacial area per unit of volume and can vary over a wide range, whereas the grain size of technical alloys is in the range of about 5–200 μm. Coating technologies such as galvanic deposition, physical vapor deposition (PVD) or chemical vapor deposition (CVD) processes lead to amorphous or nanocrystalline micro-structures possessing a high internal energy.

Two-dimensional defects affect diffusion welding in several ways: first, the dislocation movement is limited according to the grain size, since grain boundaries are obstacles for movement through the lattice. This means that for a constant strain, deformation by dislocation movement will be smaller for a material with a small grain size. At elevated temperature, however, grain growth occurs and the driving force is larger for a fine-grained material.

A similar effect is observed when using the so-called nanofoils, a thin stack of multiple nanometre layers of different materials possessing a very high interfacial energy in a meta-stable state [10, 11]. As a result, very high temperatures can be achieved temporarily.

Secondly, deformation at elevated temperatures is governed by grain boundary sliding (Coble creep) or the flow of vacancies through the volume (Nabarro-Herring creep) [12]. This means a coarsely grained material will tend to a larger deformation during diffusion welding because there are less obstacles for dislocation movement and grains tend to slide against each other.

In summary, it can be stated that the degree of deformation and the creep rate for a material during diffusion welding will depend on its grain size and will be very sensitive to the temperature used.

More complex deformation behaviour may result from multi-phase materials: phase bounda-ries can occur in a wide range of orders of magnitude, either between grains or within, e.g., as thin lamellas in grains of eutectic or eutectoid composition, such as perlite for steel.

Temporarily liquid phases (TLP) can be formed, e.g., by galvanic or PVD deposition of thin layers of two or more different metals, forming a low melting alloy during diffusion welding. Since the inter-layer diffuses into the bulk material, ideally a homogeneous material is left after finishing the process, which is insusceptible to inter-crystalline corrosion.

The opposite happens when different metals form inter-metallic compounds that are brittle and have a high melting temperature. In this case, bonding temperature and time should be limited, such that a thin layer only can be formed between both materials, which do not exhibit any excessive brittleness.

### 2.1.5. Three-dimensional defects: precipitation

As regards precipitations, it must be distinguished between soluble and insoluble species at diffusion welding temperature. Precipitation may be formed, e.g., due to a low cooling rate after diffusion welding in the range of solution annealing temperature. As a consequence, a two-phase micro-structure with coarse precipitations is formed at the grain boundaries. It is subjected to inter-crystalline corrosion (**Figure 12**). Examples are nickel-based alloys that lose their favourable corrosion resistance.
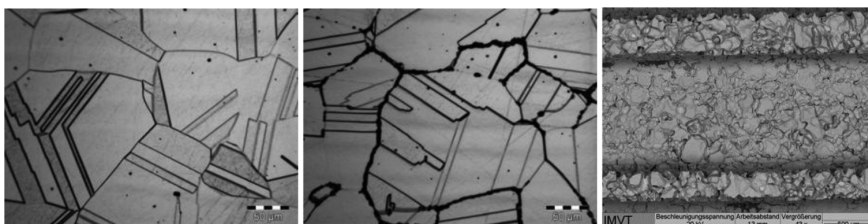
Figure 12. Micro-structure of Hastelloy C-22 (2.4602). Left: after quenching from 1100°C/70 min in water. Middle: after cooling from 1100°C with a rate of 3 K/min (1100°C ≥ 650°C = 2.5 h). Right: corrosion attack after diffusion welding in 95–97% sulphuric acid at 100°C and 1008 h.

Again, the size of precipitations determines the consequences. In case of nanoscaled, insoluble precipitations, e.g., for ODS materials, dislocation movement and grain growth is restricted very effectively [13].

For example, a pure OF-Cu showed good results at T = 850°C (**Figure 13**). The dimension of the material was 28 × 15 mm^2, consisting of six micro-structured layers with a thickness of 3.04 mm and an overall height of 13.04 mm, respectively.
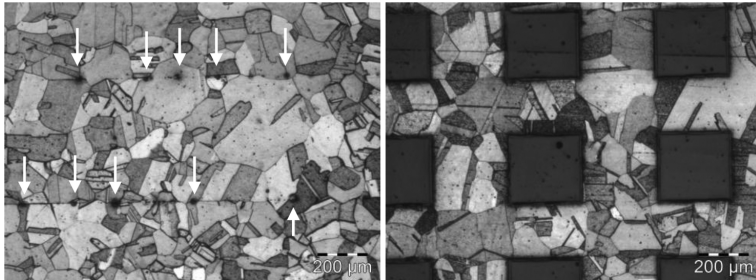
Figure 13. Diffusion welding OF copper. T = 850°C, t = 4 h, p = 2 MPa, micro-structured stack: 18.2%, overall: 4.2%.

Especially in thin-walled micro-structures, perfect grain growth across the bonding planes can be seen. However, in the massive border area, pores remain and grain growth is not as pronounced. The reason probably is a local excess of bearing pressure at the thin walls. However, in massive areas, the bearing pressure of 2 MPa is too low to deform asperities and fill pores sufficiently at this temperature.

Comparative diffusion welding experiments were made using two discs made of ODS copper Discup C3/80 with a diameter of 40 mm and an overall height of 6.88 mm (**Figure 14**). The surfaces were flycut using a polycrystalline diamond tool at a feed rate of 240 mm/min and 3000 rpm, giving a period of 80 μm feed per revolution at a very low roughness in the range of R_t = 1–1.5 μm, R_a = 0.2 μm. The roughness patterns of the discs were not aligned to each other.
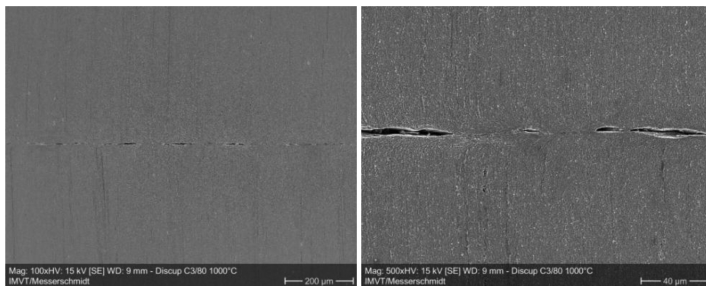
Figure 14. Diffusion welding experiment using Discup C3/80, an oxide-dispersion-strengthened copper alloy. T = 1000°C, t = 4 h, p = 6 MPa, deformation: 0.8%.

Discup alloys consist of pure copper containing a few tenths percent of sub-micron disperoids generated by reactive milling. Afterwards, the material is strongly deformed by extruding. The melting temperature is 1083°C like for pure copper. Despite a much higher temperature and bearing pressure compared to the OF copper sample shown in **Figure 13**, the deformation is as low as 0.8%. SEM images are taken, illustrating very poor joining of the mating surfaces. Grain boundaries are not visible. However, lamellar enrichment of dispersoids can be seen. Similar experiments were done using similar materials in [14].

2.2. Surface effects

For diffusion welding, a very good quality of surfaces is a pre-requisite. Surfaces must be free of single deep scratches preventing vacuum-tight joints and of impurities from machining. Careful cleaning using surfactants and subsequent rinsing with ethanol or acetone are required. Gloves free of powder should be used for handling.

The number of stacked layers will also influence deformation at the given diffusion welding parameters, since multiple surfaces are approached and levelled. Hence, it is not possible to give a certain percentage of deformation to achieve highly vacuum-tight joints. Deformation also depends on the composition of the material.

2.2.1. Influence of roughness

A pre-requisite for solid-state diffusion is a very good contact of the mating surfaces on the atomic level. Often, a "low surface roughness" that is not specified otherwise is required in the literature.

The diffusion welding process can be divided into several phases. In the beginning, surfaces are approached by the deformation of asperities. At local spots, diffusion starts on the atomic level. Between these centres, pores remain which must be closed subsequently by volume diffusion. For this, the density of vacancies and, hence, the temperature and bonding time are essential. Additionally, temperature affects the grain growth and deformation.

Several authors distinguish variable numbers of phases of the bonding process. An overview of the historical development of theoretical models can be found in [15].

Roughness influences the formation of a monolithic bond by the height and shape of asperities and the distance in between, forming temporary pores that must be filled.

Perfectly smooth surfaces made, e.g., by diamond fly cutting may prevent local deformation because asperities are lacking. Shape and height of asperities, in conjunction with bearing pressure, define the local deformation behaviour.

Asperities may also help penetrate surface passivation layers, thus producing local initial metallic contact.

2.2.2. Passivation layers

Some metals and alloys like aluminium, stainless steel, nickel-based alloys or titanium spontaneously form surface passivation layers. They consist mainly of oxides of the base metal,

some alloying elements may be enriched. Often, oxygen is blocked to prevent further oxidation and the passivation layers are responsible for the good corrosion resistance in aqueous media or hot gases. Especially for aluminium, formation of passivation layers cannot be avoided completely. The thickness of these passivation layers is in the range of 2–20 nm depending on the type of metal and the content of alloying elements [16, 17]. Of course, composition, thickness and nature of passivation layers differ for normal austenitic stainless steel, heat-resistant steels or nickel-based alloys. Hence, the diffusion welding process must be optimised and the joint must be checked for grain growth across the bonding plane (**Figure 15**). High vacuum tightness is a necessary but not a sufficient criterion.
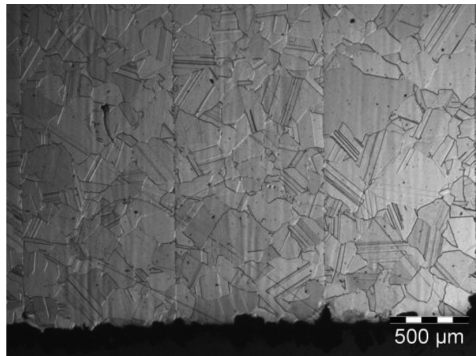

Figure 15. Diffusion-welded joint of Hastelloy C-22 (2.4602). T = 1100°C, t = 70 min, p = 12 MPa ≥ high vacuum tightness. Subsequently, solution annealed at T = 1125°C, t = 1 h, water-quenched ≥ leaky.

As mentioned above, a certain roughness may help penetrate this layer by local deformation. Hence, the passivation layer comes into contact with matrix material. Passivation layers may be removed by chemical pickling. Even if subsequent formation of a new passivation layer may not be prevented, at least a reproducible surface condition is created.

Long bonding durations and high temperatures above 80% of the starting melting temperature should be preferred in this case.

Another approach is to remove the surface passivation layer, e.g., by sputtering with argon ions. Subsequently, a layer of a different metal may be deposited, which is not that susceptible to oxidation, e.g., gold or silver, and may temporarily form a low-melting alloy helping to create a bond.

For titanium, the passivation layer is soluble in the matrix material and diffusion welding of titanium is widely used, e.g., in the aerospace industry [18, 19]. In **Figure 16**, a very good bond between thin micro-structured layers can be seen. For parts consisting of multiple thin sheets, however, it has to be kept in mind that grades 1–4 differ slightly only in terms of the contents of nitrogen, oxygen and iron, while the mechanical properties are changed dramatically [20]. Consequently, the properties of diffusion-welded parts may be changed for inappropriate ratios of surface layers to bulk material.
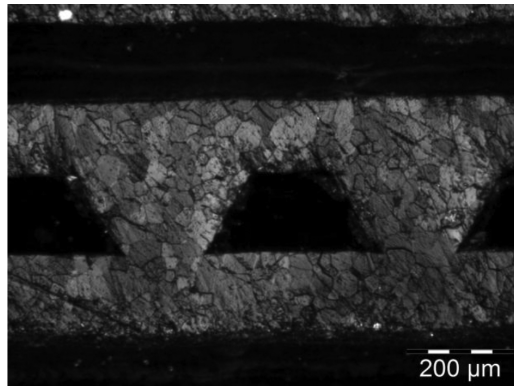
Figure 16. Diffusion weld of titanium, grade 2 at $T = 850°C$, $t = 4$ h, $p = 13$ MPa.

2.3. Influence of temperature, bearing pressure, bonding time and design

2.3.1. Influence of bonding temperature

For diffusion welding, the joining temperature is normally set in the range of about 80% of the melting temperature for a pure metal or of the starting melting temperature for alloys. Temperature is calculated in Kelvin. Obviously, similar to the appropriate temperature for recrystallization, the temperature for alloys should exceed this level. For materials with surface passivation layers, temperature should be even higher and the time longer, which changes the whole process in terms of creep rate and appropriate bearing pressure. When comparing diffusion welding of, e.g., pure aluminium ($T_s = 660°C$) and AlMg3 ($T_s = 610–640°C$), the whole process has to be optimised. Otherwise, welding will fail due to excessive deformation [21].

The influence of temperature is strongly non-linear. Keeping in mind the dependence of vacancy density on temperature, an increase of about 20 K can double the diffusion coefficient and, hence, drastically increase the creep rate for a given bearing pressure.

2.3.2. Influence of bearing pressure

Bearing pressure is responsible for joining the mating surfaces. Influence of the bearing pressure is contrary to that of temperature. When increasing the bonding time from 1 to 4 h, the deformation is not proportional but increased by a factor of about 2.5 [5].

Obviously, a certain minimum bearing pressure is necessary to facilitate the deformation of local contact areas of the sample depending on the temperature applied.

Additionally, deformation under the given conditions strongly depends on the aspect ratio of the part and on the frictional cross-section between the part and the die applying the load. Large format parts of low thickness are difficult to weld with a reproducible deformation.

If the parts contain internal thin-walled micro-structures, the deformation behaviour may be affected by grain boundary sliding. For comparison to the part displayed in **Figure 13**, a similar part with a format of 40 × 30 mm, containing 12 layers of unstructured foils with a thickness of 3.6 mm and an overall height of 13.84 mm was welded under the same conditions ($T = 850°C$, $t = 4$ h, $p = 2$ MPa). Deformation of the 12 layers was 1.3% only, while overall deformation was 0.35%, showing the influence of both micro-structures and aspect ratio.

The effective bonding area of a part should be distributed uniformly across the part. Otherwise irregular deformation or sink marks may occur (**Figure 17**). To prevent this, compensating areas may be helpful.
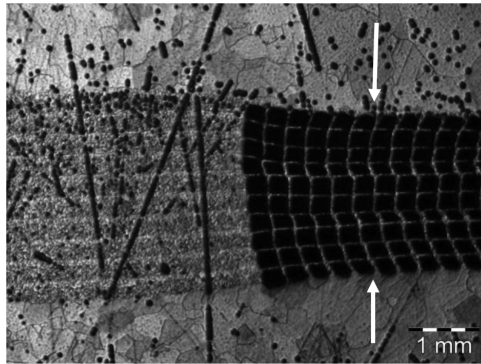
Figure 17. Irregular deformation on a part made of titanium with micro-structured sheets stacked in the same direction at $T = 850°C$, $t = 4$ h, $p = 10$ MPa.

2.3.3. Influence of bonding time

Bonding time is required for conducting the diffusion process. After the initial step of approaching mating surfaces, time is needed to fill the pores left in between the local contact areas. Hence, a sufficient long bonding time is required.

Bonding time, together with temperature, affects deformation. However, as mentioned above, its influence is non-linear. As soon as creep takes place during diffusion bonding, a long bonding time makes it difficult to control deformation and design changes may have a major impact.

Therefore, the diffusion welding process should be optimised for each serial application. It is difficult to weld prototypes of varying designs or materials without profound experience.

It is also hard to give a certain percentage of deformation to obtain a good diffusion bond. In fact, bonding quality depends on the number of layers to be bonded.

In any case, the deformation behaviour depends not only on the composition of a material but also on its micro-structure.

3. Special factors to be considered in diffusion welding of micro-devices

Some aspects relating to micro-structures have already been mentioned in the sections above. From this, it can be concluded that bearing pressure should be kept as low as possible, while, on the other hand, it must be sufficient to deform asperities and to increase the contact area during the bonding process.

The temperature should be sufficient for a high density of vacancies and for filling the pores by volume diffusion, which also depends on the bonding time.

Micro-devices mainly consist of micro-structured multiple sheets. Channels may run in the same direction or cross-wisely, and the load-bearing structures may not proceed over the whole thickness for technical reasons (**Figure 18**).
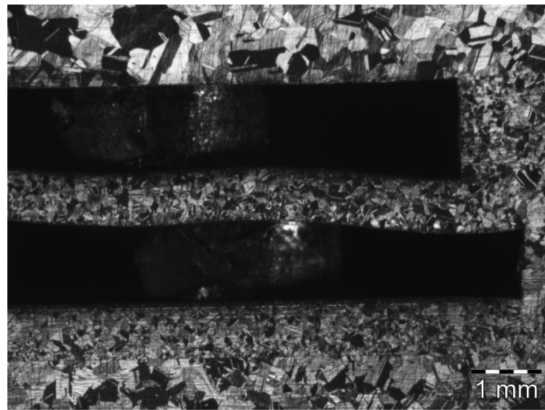

Figure 18. Displaced micro-structure with offsets made of 1.4301 diffusion-welded at locally varying bearing pressure at T = 1075°C, t = 4 h.

Bottom and top are often closed by discs of a few millimetres in thickness, having coarse grains. For thin sheet material, however, the grain size is about one order of magnitude smaller due to cold work hardening and recrystallization. The micro-structured stack and thick plates will deform completely differently and the deformation will be concentrated mostly on the micro-structured section. An intelligent design may help achieve reasonable results.

3.1. Shapes of thin walls in micro-structures

The cross-section of thin walls may be important to the deformation behaviour: if the bearing pressure forces the material to creep, cross-sections of rectangular wall may be bent or deformed to a barrel shape, as can be seen in the left section of **Figure 19**. In the SEM of material with etched micro-channels on the right, however, the part is stabilised, since the bonding cross-section increases when deformation occurs.
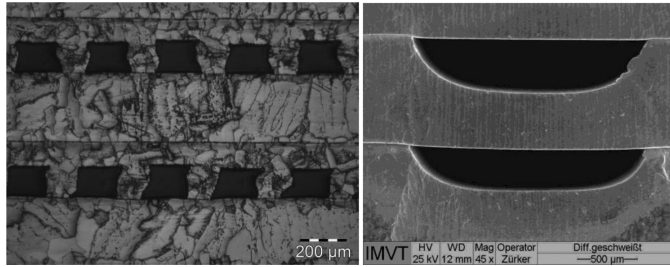
Figure 19. Impact of the cross-section of thin walls. Left: rectangular cross-section of thin walls. Right: self-reinforcing cross-section due to etched micro-channels.

The dimension of the walls should exceed the grain size of the material: in most cases, walls should be at least 100–200 μm in width. The aspect ratio should not exceed 1:1 for stability reasons, e.g., to avoid bending.

3.2. Impact of the design of mechanical micro-structures, the aspect ratio and the number of layers on the deformation

Moreover, the geometry of the micro-structured foils to be proper is important: the ratio between the thickness of the remaining bottom and the width of a trench should not exceed 1:1 to transmit sufficient bearing pressure to the next layer and to prevent lacking fusion (**Figure 20**, left).
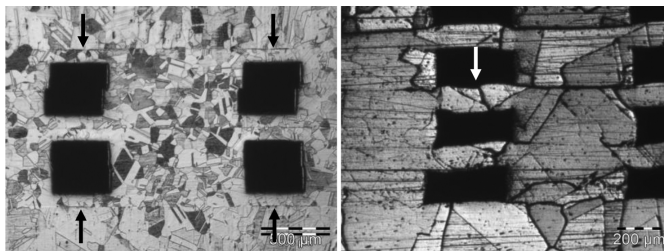
Figure 20. Left: 1.4301, T = 1075°C, t = 4 h, p = 8 MPa, incomplete fusion due to insufficient thickness of the bottom in relation to the width of trenches. Right: 1.4876, T = 1250°C, t = 1 h, p = 8 MPa, distortion due to grain boundary sliding within a thin bottom.

Depending on the application, a grain boundary crossing the remaining thickness of the bottom of a micro-channel should be avoided. In case of corrosion, this would be a favourable path for failure. During diffusion welding it causes local grain boundary sliding and distortion of the mechanical micro-structure (**Figure 20**, right).

Another topic is the aspect ratio of the parts to be welded, e.g., due to the different thermal expansion coefficients of the TZM-stamps (see Section 4), and the parts and deformation during the welding process, friction between both occurs. For a high aspect ratio in the range

of one or more, a barrel-shaped profile results, accompanied by a high percentaged deformation. Flat parts, however, possess a low deformation at the same conditions. For example, for disks of 160 mm in diameter, a deformation of 10% was obtained for a height of 10 mm for T = 1075°C, t = 4 h, p = 25 MPa. For a height of 150 mm, however, the deformation was more than 33% [22].

The number of layers affects the deformation obtained at the same conditions since the roughness of more surfaces must be levelled. For example, a conical sample consisting of 51 layers had a deformation more than 30% higher than the same sample geometry consisting of five segments only (**Figure 21**).

Figure 21. Conical samples made of 1.4301, T = 1000°C, t = 4 h, F = 17.55 kN, corresponding to 15–25 MPa. Left: Before diffusion welding. Middle: Five segments, deformation: 5.41 and 5.11%, respectively. Right: Sample made of 51 layers; deformation: 8.34% [6].

4. Equipment for diffusion welding

Diffusion bonding can be carried out using hot isostatic pressing (HIP) at a high isostatic pressure applied by argon of up to 2.500 bar or using a heated press with uniaxial load. For HIP, the parts must be placed inside a steel shield container which is evacuated before sealing. This makes the handling of the parts and the process itself rather expensive.

Additionally, sticking of the parts to the container must be prevented, e.g., by rock wool layers in between or boron nitride spray, or the container has to be machined off afterwards. When using fibrous materials, desorption from a high specific surface area at high temperatures has to be considered. However, also parts with an irregular bonding plane can be welded by HIP, since homogeneous pressure is applied. HIP is widespread and offered by service providers, e.g., ABRA Fluid AG [23].

Diffusion bonding using uniaxial heated presses is performed under a protective inert gas atmosphere or high vacuum. Only a few companies supply equipment for diffusion welding,

e.g., PVA TePla AG, FormTech GmbH, TAV VACUUM FURNACES SPA and Centorr Vacuum Industries. Other companies such as MAYTEC Mess- und Regeltechnik GmbH and SYSTEC Vacuum Systems GmbH & Co.KG modify equipment like tensile testing machines or produce equipment for special needs (**Figure 22**). For this, a water-cooled vessel with a vacuum-sealed feedthrough for the dies is installed. The oven is heated indirectly by metallic heaters, and a vacuum in the order of 1E-05 Pa must be maintained for the protection of the heaters. Temperatures of not more than 1400°C are sufficient for the most commonly used materials.

Figure 22. Diffusion bonding furnaces. Left: Maytec diffusion bonding furnace, maximum force 20 kN. Right: Systec diffusion bonding furnace, maximum force: 2 MN.

The stamps are often made of TZM, a molybdenum ODS-alloy, possessing still a high mechanical stiffness at high temperatures [24]. However, the stability also depends on the thickness-to-diameter ratio and must be adapted to the forces transferred to the sample to prevent irregular deformation of the parts to be welded.

Due to the thermal mass of the equipment and to limit thermal stress, the heating rate and especially the cooling rate are low. PVA TePla AG also offers a rapid cooling technology for decreasing the cycle time [25].

During diffusion welding of stainless steel and nickel-based alloys under vacuum, chromium depletion takes place at the surface due to high partial pressure of chromium oxide [26, 27]. Hence, corrosion properties differ from a heat treatment in inert gas or air. For these materials, also enrichment of carbon must be prevented. Hence, unshielded heaters made of graphite are unsuitable.

5. Discussion and outlook

Diffusion welding is the only welding process allowing for full cross-sectional welding, mostly without any liquid phase formation. Since the whole part is subjected to a heat treatment, attention must be paid to undesired material changes. Any cold work hardening effect disappears and the grain size will be larger than before.

With reasonable efforts, high-melting metals, e.g., tungsten or tantalum, cannot be welded.

The equipment is expensive. The process mostly runs batch-wise. Depending on the machinery and the geometry of the parts to be joined, the output is relatively low. Mostly, costs are high.

The process has to be optimised with respect to temperature, bearing pressure and time, taking into account the composition of an alloy and the mechanical history of the semi-finished product. This makes it an interesting field of research for materials scientists. High temperatures and long bonding times are favourable as long as grain growth is not important.

Overall, the result of diffusion welding is difficult to control and depends on many other geometrical factors as well. Therefore, it is used mainly for special applications or in the aerospace industry where cost pressure is lower.

The design of a part must be adapted to diffusion welding, e.g., in terms of a constant distribution of the bonding net cross-section across the part to prevent sink marks. High vacuum tightness is a necessary but not a sufficient criterion for diffusion welding of apparatuses.

To obtain good welding results, a certain deformation always must be accepted. It depends on, e.g., the aspect ratio of the parts and the number of layers to be joined. Obviously, micro-channels inside a part will affect the amount of deformation additionally. For multi-layered parts, a higher deformation is required to achieve high vacuum tightness since more surfaces have to be levelled. In consequence, it needs a lot of experience to define appropriate parameters, especially for the bearing pressure, to ensure a sufficient deformation related to the number of layers. Hence, it is not possible to give an exact value of deformation necessary to obtain high vacuum tightness for a material itself.

Since a long bonding time makes it more difficult to control the deformation at a constant bearing pressure, a short increase of bearing pressure for approaching the surfaces may be helpful. Time should be given in between for closure of remaining pores at a reduced constant bearing pressure without a steady strain rate.

As shown for different types of steel, also the material properties and surface passivation layers may have high impact on the behaviour during diffusion welding. Not all materials of the same class can be welded at the same temperature since passivation layers may possess different thermal stability. Often an increased temperature is required to achieve grain growth across the bonding planes, depending on the alloying elements and its content.

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