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Engineering Physics and Mathematics

CREEP EFFECTS IN DIFFUSION BONDING OF OXYGEN-FREE COPPER

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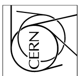

BACHELOR'S THESIS

17.4.2013

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AALTO UNIVERSITY SCHOOL OF SCIENCE PO Box 11000, FI-00076 AALTO http://www.aalto.fi	ABSTRACT OF THE BACHELOR'S THESIS

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Abstract:

Diffusion is the transport of atoms or particles through the surrounding material. Various microstructural changes in metals are based on the diffusion phenomena. In solid metals the diffusion is closely related to crystallographic defects. In single-component metals the dominant mechanism of diffusion is the vacancy mechanism.

Diffusion bonding is a direct technological application of diffusion. It is an advanced solid-state joining process in which the surfaces of two components are brought to contact with each other and heated under a pressing load in a controlled environment. During the process, the contact surfaces are bonded by atomic diffusion across the interface and as a result, one solid piece is formed. The condition of high temperature and low applied stress combined with relatively long process duration enables the creep effects to take place in bonded metals. Furthermore, creep causes unwanted permanent deformations in the bonded components. Some authors suggest that there could be a threshold for stress below which the creep mechanisms do not activate.

In case study, an experimental test on diffusion bonding of the CLIC accelerator structure disks conducted in CERN is introduced with a corresponding finite element simulation. Also a comparison between the results of the experimental test and the simulation is presented. The accelerator structure disk material is oxygen-free electronic (OFE) copper.

The observed permanent deformations of the disks in the experimental test were very low or negligible but the bonding joint was partially defected. In turn, some deformations were observed in the simulation. It is possible that the used bonding pressure was just below a threshold stress. In the finite element simulation the creep strain-rates are calculated even for the lowest stresses, which could explain the observed deformations. Based on the results of this and previous experimental tests conducted in CERN it can be concluded that the threshold stress for creep exists and that the value of the threshold is around 0.05 MPa in this case.

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Tiivistelmä:

Diffuusiolla tarkoitetaan atomien tai partikkelien liikettä ympäröivässä materiaalissa. Monet metallien mikrorakenteen muutokset perustuvat diffuusioilmiöön. Kiinteässä metallissa diffuusio perustuu metallin kiderakenteen virheisiin. Yleisin diffuusiomekanismi puhtaissa metalleissa on vakanssi- eli korvaussijadiffuusio.

Diffuusioliittäminen on diffuusion suora teollinen sovellus. Diffuusioliittäminen on pitkälle kehitetty kiinteän olomuodon liittämismenetelmä, jossa kahden metallikappaleen pinnat tuodaan lähelle toisiaan, ja kappaleita kuumennetaan puristavan voiman alaisena kontrolloidussa ympäristössä. Prosessin aikana metallikappaleet liittyvät yhteen rajapinnalla tapahtuvan diffuusion vaikutuksesta. Diffuusioliittämisessä tapahtuva suhteellisen pitkäkestoinen altistus korkealle lämpötilalle sekä pieni puristusjännitys aiheuttavat metalleissa virumista, joka taas saa aikaan ei-toivottuja pysyviä muodonmuutoksia yhteenliitettävissä kappaleissa. Jotkut tutkijat ovat ehdottaneet, että jännitykselle olisi olemassa kynnysarvo, jonka alapuolella virumismekanismit eivät aktivoidu.

Tapaustutkimuksessa esitellään CERN:ssä toteutettu kokeellinen tutkimus, jossa liitettiin yhteen CLIC-hiukkaskiihdyttimessä käytettäviä superjohtavasta kuparista (OFE-kupari) valmistettuja kiekkoja diffuusioliittämisen avulla. Lisäksi esitellään vastaava elementtimenetelmäsimulaatio sekä vertaillaan simulaation ja kokeellisen tutkimuksen tuloksia.

Kokeellisessa tutkimuksessa havaitut pysyvät muodonmuutokset kiekoissa olivat hyvin pieniä tai olemattomia, mutta diffuusioliitos oli osittain puutteellinen. Simulaatiossa puolestaan havaittiin jonkin verran pysyviä muodonmuutoksia. On mahdollista, että diffuusioliittämisessä käytetty puristusjännitys oli juuri kynnysarvon alapuolella. Simulaatiossa virumisesta aiheutuvat venymät lasketaan myös pienimmille jännityksen arvoille, mikä saattaa selittää havaitut muodonmuutokset. Tämän testin ja aikaisempien CERN:ssä toteutettujen kokeellisten tutkimusten perusteella voidaan päätellä, että virumiselle on olemassa kynnysarvo, joka tässä tapauksessa on noin 0.05 MPa.

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Avainsanat: diffuusio, diffuusioliittäminen, viruminen, hapeton kupari, elementtimenetelmä

FOREWORD

This Bachelor's Thesis has been made during the spring 2013 in collaboration between Aalto University, CERN, and Helsinki Institute of Physics (HIP).

I would like to express my gratitude to Kenneth Österberg (HIP) and Germana Riddone (CERN) for the opportunity to work in CERN during the summer 2012. I am also grateful for Prof. Mikko Alava (Aalto University) for his guidance and for Jukka Väinölä (HIP, CERN) for his valuable comments during the work. Great thanks to D.Sc. (Tech.) Fabrizio Rossi (CERN) who also reviewed the work as well as to the whole CLIC team for the help and guidance in my work during the summer 2012.

Finally, I thank Hanna for her support and patience during this thesis work.

Oulu, April 17, 2013

Author

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1 INTRODUCTION

The base work for this Bachelor's Thesis has been done during summer 2012 in the European Organization for Nuclear Research (CERN) in the framework of Compact Linear Collider -project (CLIC). The ongoing CLIC project is aiming to design and build a next generation particle accelerator. At the moment CERN is running the world's largest and highest-energy particle accelerator Large Hadron Collider (LHC). After its life cycle comes to an end, there will be a need for a new particle accelerator; approximately after year 2020. With CLIC a new energy region could be explored which may stay unreached by the LHC. This would allow the detection of new particles and testing of different models of modern physics. (Aicheler *et al.*, 2012)

The accelerating structures of CLIC are assembled using brazing and diffusion bonding as the main joining processes. During diffusion bonding, joined components undergo elastic and plastic deformations due to high temperature and low pressure between contacting surfaces. With current parameters, the material of CLIC accelerating structure disks is high conductivity oxygen-free electronic (OFE) copper. Accelerating structures are exposed to very high gradient electric and magnetic fields that require ultra-low operating temperature. OFE copper has high thermal and electrical conductivity and it is practically pure metal with the oxygen level of 0.001 % or below. Alloying and impurities would decrease the electrical conductivity, and therefore OFE copper is the standard material for radio frequency applications such as the particle accelerator structures. (Aicheler, 2012, ASM Handbook, 1992)

In order to get adequate control over the diffusion bonding process, it is necessary to understand the thermo-mechanical deformations due to diffusion bonding. According to the state-of-the-art studies, the main cause of deformations in diffusion bonding at high temperature and low pressure is diffusional creep. Moreover, the physical background of creep lies in the diffusion phenomena. Consequently, the basics of diffusion theory and main mechanisms of diffusion in pure copper are presented in the chapter 2. Furthermore, diffusion bonding and its process variables are explained in the chapter 3. After that the definition and basic mechanisms of diffusional creep are introduced in the chapter 4.

Within tight manufacturing tolerances, it is reasonable to optimize the bonding process parameters so that the permanent deformations of the accelerating structure disks due to the assembling are reduced as low as possible. However, conducting a series of experimental bonding tests for optimization would be unnecessarily expensive and time-consuming. Thus, preliminary process optimization is reasonable to accomplish with a computer simulation. In the chapter 5, a case study including a finite element simulation of diffusion bonding process of accelerating structure disks is introduced. Also a comparison with recently conducted experimental tests is presented.

This thesis work does not go deep into the mathematical formulation of diffusion or creep but concentrates on describing the phenomena in an understandable way regardless of the reader's initial knowledge of the topic. Only the basic creep formulas and diffusion laws are presented in this work.

2 DIFFUSION IN PURE METALS

Diffusion is defined as the transport of atoms or particles through the surrounding material. Diffusion occurs in gases and liquids as well as in amorphous and crystalline solids. In metals, the kinetics of various microstructural changes base on the diffusion phenomena. Diffusion is strongly present in, for example, preparation, processing, and heat treatment of metals. (Mehrer, 2007, Ashby & Jones, 1980: 180-185) In this chapter, the principles of the most common diffusion mechanisms in pure metals are introduced.

2.1 Lattice structure

In a solid metal, the atoms are packed together forming three dimensional crystal structures, *lattices*. Lattice structures give the minimum of total bonding energy, being the most likely assembly of the atoms. Metal lattices are given descriptive names by the arrangement of the atoms in the structure. The most common lattices are body-centered cubic (BCC), face-centered cubic (FCC), and hexagonal close-packed (HCP) structure. In BCC the atoms are arranged at the corners of the cube with another atom at the center of the cube center. BCC lattice structure is presented in the figure 1. Examples of metals having BCC crystal structure at room temperature are tungsten and molybdenum. (Kazakov, 1981: 18, Ashby & Jones, 1980: 47-51, 180-185)

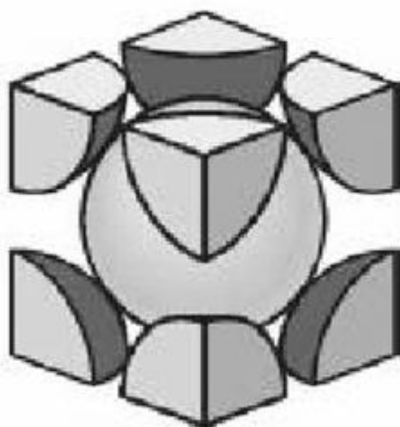

Figure 1. Body-centered cubic (BCC) lattice structure (Monteiro, 2012).

In FCC the atoms are located at each of the corners and the centers of all the cubic faces: see figure 2. FCC is the lattice structure of copper and, for example, aluminium. HCP is very similar to the FCC structure, but instead of being a cubic structure, the pattern is hexagonal. HCP lattice structure is presented in the figure 3. Magnesium and titanium are examples of metals having HCP lattice structure at room temperature. (Kazakov, 1981: 18, Ashby & Jones, 1980: 47-51, 180-185)

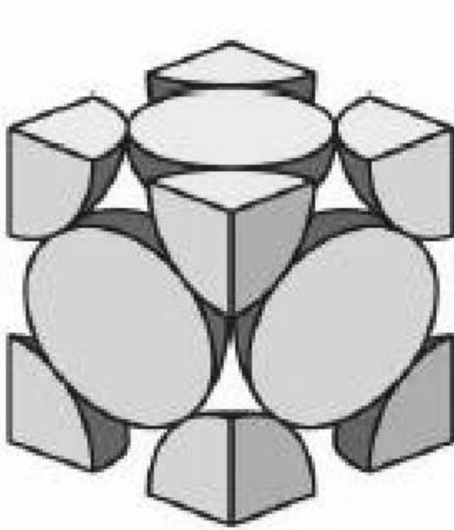

Figure 2. Face-centered cubic (FCC) lattice structure (Monteiro, 2012).

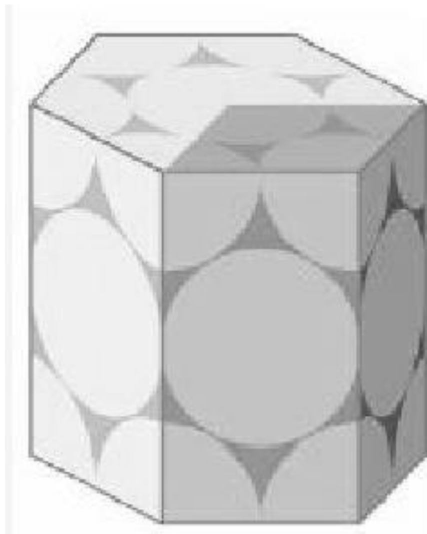

Figure 3. Hexagonal close-packed (HCP) lattice structure (Monteiro, 2012).

Crystal lattices define the mean positions of atoms in solid metals. As atoms oscillate around their mean positions, they continuously collide to each other transferring energy. With a certain probability, an atom can obtain enough energy to “jump” from an atomic site to another. In specific words, the amount of energy is sufficient for an atom to overcome the energy barrier between two adjacent atomic sites. This energy barrier is called the *activation energy*. Jumping atoms leave behind unfilled bonds in the original lattice site. These vacant lattice spaces are called the *vacancies*, which, among other lattice imperfections, are discussed next. An illustration of lattice structure with a vacancy is presented in the figure 4. (Mehrer, 2007: 68-80, Ashby & Jones, 1980: 180-183)

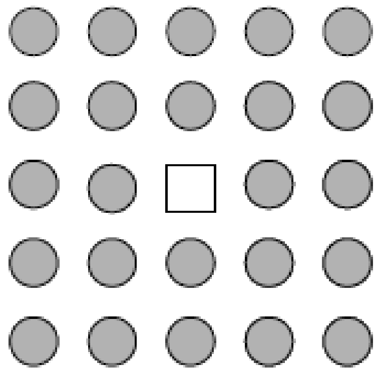

Figure 4. Lattice structure with a vacancy (Mehrer, 2007: 136).

2.2 Lattice imperfections

In practical terms, lattices are never completely pure and organized. There occur different kinds of local and structural crystallographic defects in the lattice, for example, point defects and line defects. Besides the already mentioned vacant atomic sites, there can be “odd” atoms between the lattice sites that are called the *interstitial atoms*. Interstitial can be either an extra atom located between the lattice sites in pure metal (*self-interstitials*) or a foreign solute atom such as hydrogen (H), nitrogen (N), carbon (C), or oxygen (O). Both the vacancies and the interstitial atoms are classified as point defects due to their physical dimensions and localness. In close-packed cubic metals as copper the vacancies are more common type of point defects than the self-interstitials. This is due to higher formation energy of self-interstitials than that of vacancies.

(Mehrer, 2007: 69-100, Ashby & Jones, 1980: 95-101) Illustrations of lattice structures with interstitial atom and self-interstitial are presented in the figures 5 and 6.

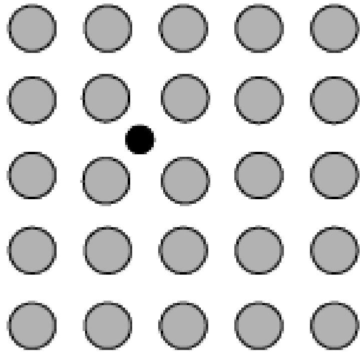

Figure 5. Lattice structure with an interstitial atom (Mehrer, 2007: 137).

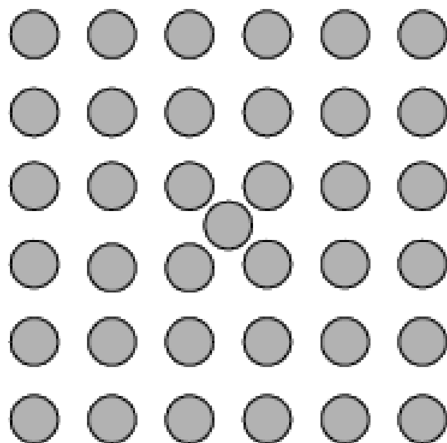

Figure 6. Lattice structure with a self-interstitial (Mehrer, 2007: 137).

Line defects are often referred as the *dislocations*, which are particular types of structural defects that can cause the material to deform plastically at relatively low stresses. Edge and screw dislocations are examples of such defects that can produce plastic strain with relative ease when they are moving between the well-organized crystal lattice structures. *Grain boundaries*, *phase boundaries*, and *free surfaces* are other examples of structural defects in solid metals. The mobility of atoms is usually considerably higher along these defects and dislocations than straight through the crystal lattice by diffusional flow. (Mehrer, 2007, Ashby & Jones, 1980: 95-101)

2.3 Diffusion mechanisms

There are various mechanisms of diffusion depending on the materials that are involved in the diffusion process. Different mechanisms of diffusion are, for example, interstitial, collective, vacancy, and interstitial-substitutional exchange mechanism. However, as many of these mechanisms are related to solute atom diffusion, only couple of different mechanisms of diffusion should be considered for pure metals. In single-component metals the diffusional flow of material is generally called the *self-diffusion*. (Mehrer, 2007: 94-103)

Mechanisms of diffusion in metals are closely related to the lattice point defects. Two basic mechanisms of diffusion are the *interstitial mechanism* and the *vacancy mechanism*. In pure metals, the vacancy mechanism is the most probable mechanism of diffusion, while the interstitial mechanism is the most probable in case of impurities or solute atoms are involved in the diffusion process. For alloys the diffusion mechanisms are more complex than for single-component metals. In this thesis the possible diffusion mechanisms are considered only for pure metals, or more specifically, for pure copper. Although the vacancy mechanism is the most common diffusion mechanism in pure metals, also the interstitial mechanism is presented in this context since it may occur in pure metals by the *interstitialcy mechanism* related to self-interstitial atoms that are described below. Also, the interstitial mechanism is useful for removing oxides and other impurities from metals that are required to maintain their original purity during, for example, heat treatment. (Mehrer, 2007: 94-103, Kazakov, 1981: 21-24) In addition, some short circuit diffusion mechanisms are introduced at the end of this chapter.

2.3.1 Interstitial mechanism

Unattached, the interstitial atoms have more energy than the atoms in the equilibrium crystal lattice sites. Since the atom system is always seeking lowest energy state, any loose atoms moving within the range of interatomic forces can be easily captured by unfilled bonds (vacancies or interstitial sites) in the lattice. This is the basic mechanism of interstitial atomic movement leading to material flow through the lattice. (Kazakov, 1981: 18-24, Ashby & Jones, 1980: 180-185)

Interstitial movement is the most probable mechanism of diffusion for foreign solute elements with a small atomic radius like O, H, N, and C. These kind of small atoms diffuse through the crystal lattice very rapidly. (Mehrer, 2007: 94-96, Ashby & Jones, 1980: 185) The basic idea of interstitial mechanism is presented in the figure 7. Gray spots describe the lattice atoms and the black spot describes the interstitial atom, whereas the black arrows represent the interstitial movement path. (Mehrer, 2007: 96)

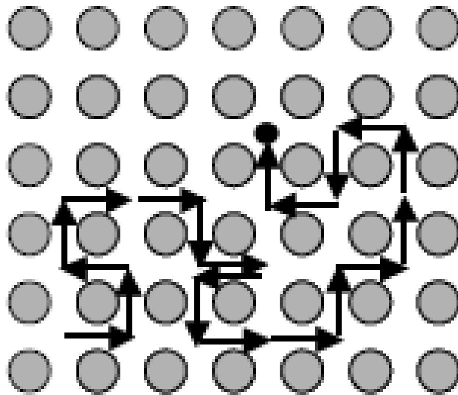

Figure 7. Interstitial mechanism of diffusion (Mehrer, 2007: 96).

2.3.2 Interstitialcy mechanism

In pure metals, self-interstitials require a great amount of energy to jump from an interstitial site to another, since the atom has to migrate through a pair of tightly bonded atoms equal in size to the self-interstitial. Usually, it is necessary for another atom or atoms to move simultaneously with the self-interstitial in order to even some movement to happen. This mechanism is called the interstitialcy mechanism and it is quite rarely seen as a direct atom exchange mechanism in pure metals. In fact, it is said to be negligible mechanism for thermal diffusion of metals due to the high formation enthalpies of self-interstitials. (Mehrer, 2007: 100-102, Kazakov, 1981: 21-24)

However, the exchange of atoms in pure metals occurs more readily if multiple atoms get involved in the exchange process simultaneously. These kinds of diffusion mechanisms are referred as the *collective mechanisms*. For example, in ring mechanism three or more atoms rotate as a group by the distance of one atom. Still, it has been

shown that these non-defect mechanisms of diffusion are more common in amorphous systems than in crystalline solids. (Mehrer, 2007: 97-98)

2.3.3 Vacancy mechanism

The exchange of pure metal atoms is accelerated significantly, if there are vacancies in the lattice. Consequently, the dominant diffusion type in pure metals and alloys is the vacancy mechanism. In this mechanism, a vacancy in the lattice changes place with one of the adjacent atoms, and then this process is repeated again and again. As a result, a flow of atoms occurs to the opposite direction than that of the vacancies. The activation energy for vacancy mechanism is lower than for interstitial or collective mechanisms. (Kazakov, 1981: 18-24, Mehrer, 2007: 98-100, Butrymowicz *et al.*, 1973) An illustration of the vacancy diffusion is showed in the figure 8. White square represents the vacancy, grey spots are the matrix atoms, and black spot is the jumping atom.

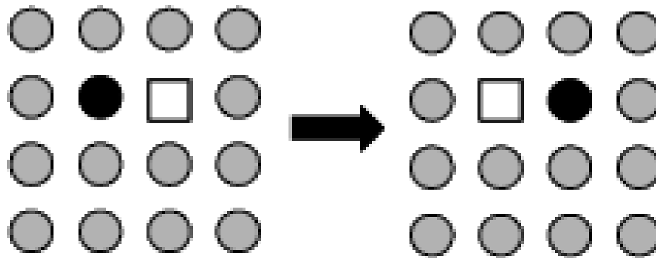

Figure 8. Vacancy mechanism of diffusion (Mehrer, 2007: 99).

Vacancies can be filled when one of the adjacent atoms has enough energy to jump into the vacant lattice site. At the same time, energy is needed for vacancy formation as interatomic bonds must be broken. Therefore, the total activation energy is defined as the sum of the energies required by these two processes. (Kazakov, 1981: 18-24, Mehrer, 2007: 98-100, 300)

At temperatures higher than about 2/3 of the melting temperature, two or more vacancies can gather around making it easier for an atom to jump into one of the adjacent vacancies. These aggregates of vacancies are called divacancies (or trivacancies and so on) and the diffusion mechanism is called, respectively, the

divacancy mechanism. This leads to a higher diffusion rate than in the case of single vacancy mechanism. (Mehrer, 2007: 100)

2.3.4 Short-circuit mechanisms

Diffusion in solid metals may be short-circuited by diffusion via structural defects like grain boundaries, free surfaces, and dislocations that were discussed already earlier. These mechanisms of diffusion are considerable in case there are lots of dislocations in the metal, or when the average grain size is small. Furthermore, short-circuit diffusion is the dominant mechanism at low temperatures due to its low activation energy compared to that of vacancy diffusion. (Ashby & Jones, 1980: 186, Butrymowicz *et al.*, 1973) Illustrations of two kinds of fast diffusion paths are presented in the figures 9 and 10.

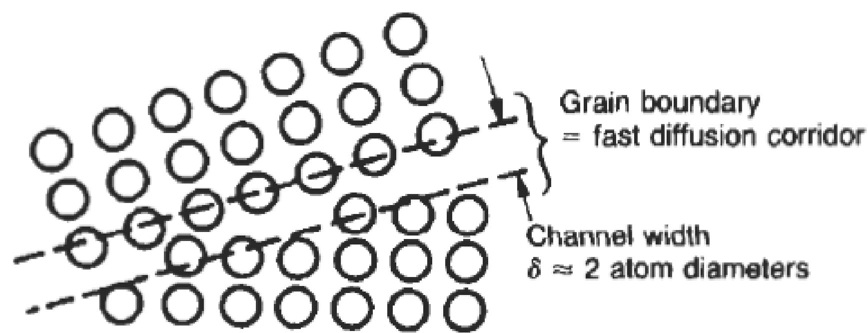

Figure 9. Fast diffusion path via grain boundary (Ashby & Jones, 1980: 186).

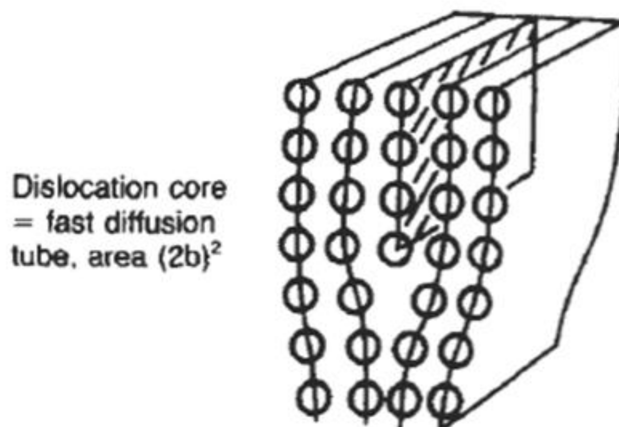

Figure 10. Fast diffusion path via free surface (Ashby & Jones, 1980: 186).

3 DIFFUSION BONDING PROCESS

Diffusion bonding is a direct technological application of the diffusion phenomena. In this chapter, an introduction to diffusion bonding process and its properties is presented. A case study of diffusion bonding to the Compact Linear Collider –project at CERN is presented in the end of thesis including a description of experimental bonding tests and a finite element simulation of the bonding process.

3.1 Definition of diffusion bonding

Diffusion bonding is an advanced solid-state joining process in which the surfaces of the components to be joined are brought to contact with each other and heated under a pressing load in a controlled environment. During the process, contact surfaces are bonded by local plastic deformations and atomic diffusion across the interface at elevated temperature and as a result, one solid piece is formed. By diffusion bonding a joint can be made between similar and dissimilar metals and alloys as well as non-metallic materials that would be difficult or impossible to join by conventional welding processes. (Kazakov, 1981: 9-16) A schematic illustration of diffusion bonding process in which two copper disks are joined together is presented in the figure 11.

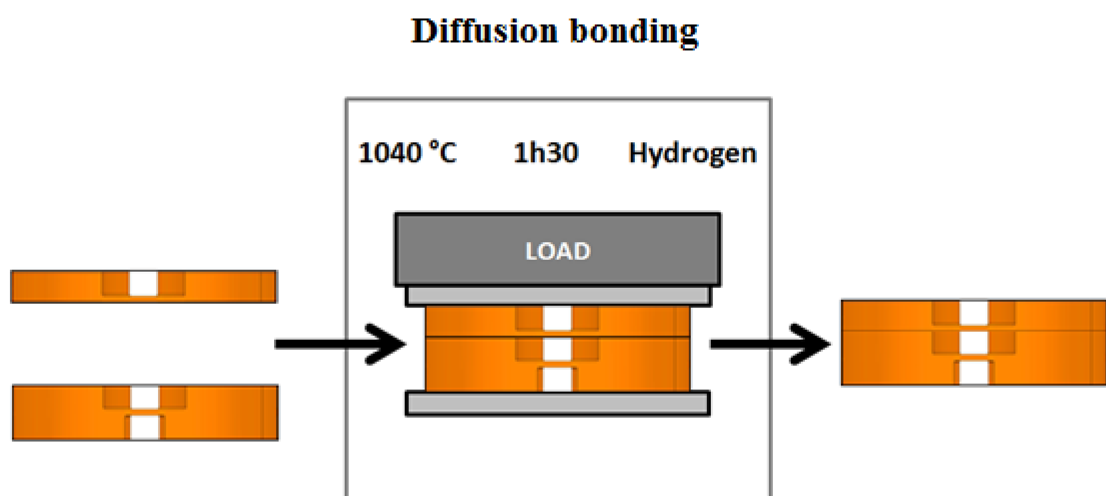

Figure 11. A schematic illustration of joining together copper disks by diffusion bonding. (courtesy of CERN)

3.2 Process variables

The success of the diffusion bonding process depends mainly on three adjustable variables: the bonding temperature, the applied pressure or pressing load, and the holding time. The quality of the achieved bonding joint depends also on the bonding environment (for example vacuum or reducing atmosphere), the material properties, and the preparation of the contact surfaces. With appropriate process variables and setup, the bonding joint will have the same tensile strength and ductility as the parent material. (Kazakov, 1981: 9-16)

3.2.1 Bonding temperature

The *bonding temperature* is usually between 0.5 and 0.7 of the absolute melting point of the most fusible material in the bonded composition. Elevated temperature aids the plastic deformation of surface asperities and accelerates the atomic diffusion across the bonding interface. In some applications bonding temperature can be even higher. Then, only low applied pressure is needed to achieve a sound joint. (Kazakov, 1981: 11-60)

3.2.2 Bonding pressure

To avoid any unwanted macroscopic deformations, the applied *bonding pressure* is usually kept as low as possible, yet achieving a sufficient bonding joint. The components are compressed together with a force able to disperse the oxide films on the contact surfaces. Even though applied stress is generally below the yield strength of parent material, the yield point can be exceeded at the peaks of surface asperities. Plastic deformation of the machining ridges and surface asperities enables the surface crystals to reach contact within an atomic distance, which is an important precondition for the diffusion mechanisms to take place. Sufficient bonding pressure also ensures that all gaps and voids are filled at the interface during bonding. (Kazakov, 1981: 11-60)

3.2.3 Holding time

The *holding time* at the effective bonding temperature and pressure is also reasonable to keep to a minimum with a limitation that it still has to be long enough for the diffusion processes to take place. Bonding for excessive time can leave voids in the joint or lead

to unwanted changes in the chemical composition of the metals. Also, shorter holding time is more sustainable in terms of energy consumption, which makes it economically and environmentally profitable. Controlling and optimizing the holding time is very important especially in mass production applications of the diffusion bonding. (Kazakov, 1981: 11-13, 53-60, Aicheler *et al.*, 2012)

3.2.4 Bonding environment

The *bonding environment* effects on the joint quality. A vacuum cleans the surfaces and sublimes the oxide films improving the material properties at the diffusion zone. Similar reaction can be achieved by a reducing atmosphere as hydrogen. Sometimes an inert gas is used as a medium in the bonding process chamber. (Kazakov, 1981: 11-13, 53-60)

3.2.5 Bonding heating cycle example

A real bonding temperature curve measured in experimental bonding tests done at Bodycote (Annecy, France) in December 2011 by Fabrizio Rossi (CERN) is presented in the figure 12. Temperature of the bonded components is measured using thermocouples. The heating cycle includes several phases that can be tuned according to the facilities used, safety requirements, and so on. First, high vacuum up to 10^{-5} mbar is generated in order to clean the furnace atmosphere. Secondly, insertion of hydrogen at partial pressure of around 20 - 35 mbar is followed by heating of the components up to 1025 °C. After diffusion bonding in hydrogen at 1025 °C for 1.5 hours, slow cooling is started and at around 750 °C the hydrogen is removed and replaced with an inert shielding gas as argon. Finally, the insertion of the argon is followed by fast cooling. (Rossi, 2011 and 2012)

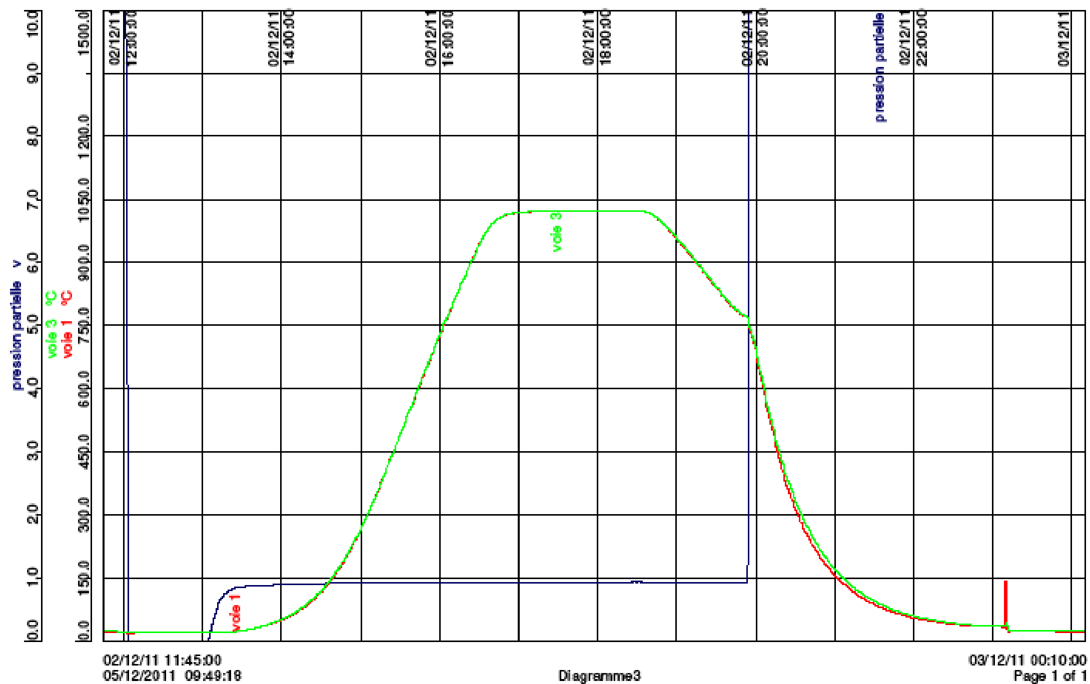

Figure 12. An example of bonding temperature curve. (Rossi, 2011)

3.3 Microstructural considerations

Traditional theoretical approach in literature is that there are two main phases in diffusion bonding. Firstly, a close contact between the surfaces is achieved by deformation and diffusive leveling of surface asperities and possible machining ridges. Secondly, the bonding joint is formed by the formation of atomic bonds and closure of voids due to diffusion mechanisms. Over time, the joint is expanded into the parent materials as stress relaxation and reduction in defect concentration takes place in the bonded zone. The wider the bonded zone gets, the stronger and more reliable the resulting joint will be. After a successful diffusion bonding, there should be crossing grains along the bonding interface. (Kazakov, 1981: 17-26, 49-56)

During diffusion bonding process metals undergo several microstructural changes as grain growth, recrystallization, and recovery processes. The changes are mainly caused by the elevated temperature combined with long process duration, but also the applied pressure affects the microstructure. (Kazakov, 1981: 49-60, Aicheler *et al.*, 2011) All of these microstructural changes are important in the formation of bonding joint. However,

in this thesis the main subject is creep or more specifically the creep effects related to diffusion bonding. Thus, assuming as the most important microstructural considerations related to the subject, grain growth and material behaviour are discussed next.

3.3.1 Grain growth in diffusion bonding

It is a well-established concept in metallurgy that grain size increases as metals are heated or kept at elevated temperature for long time. The influence of time and temperature on grain size is demonstrated generally in the figure 13. The curve represents the grain size at certain points of temperature and holding time. It can be seen from the figure that even at relatively low temperatures with holding time of few hours, the grain size can multiply tenfold. (MET, 2001: 75-80, Kazakov, 1981: 54-60)

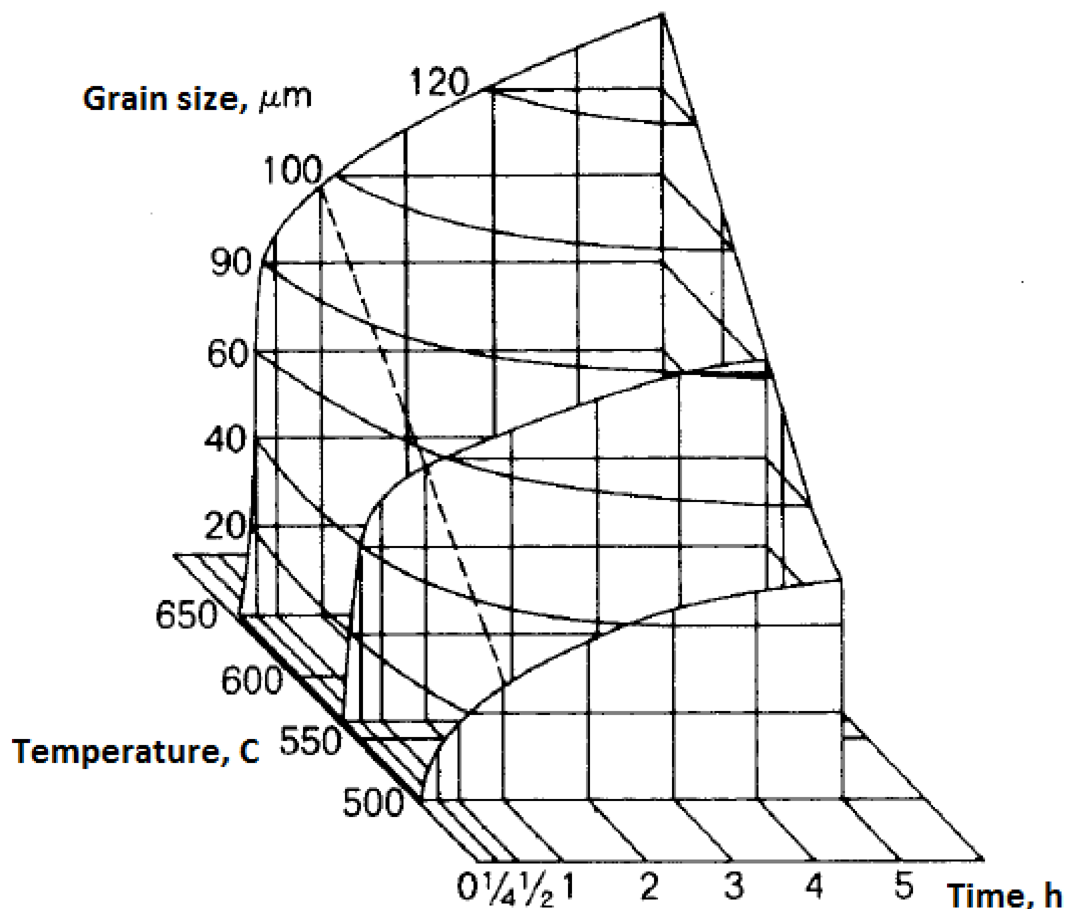

Figure 13. Grain size as a function of time and temperature. Translation from the original picture (MET, 2001: 77).

Also in diffusion bonding, an excessive holding time or high bonding temperature can lead to a rapid grain growth (Kazakov, 1981: 54-60). It has been experimentally observed that impurities and solute atoms lower the mobility of grain boundary dislocations decreasing the grain growth. In turn, extensive and abnormal grain growth occurs in pure metals at high temperatures. (Wilshire & Palmer, 2001, Frost & Ashby, 1982: 128)

In the heat cycle experiment conducted in CERN, Aicheler (*et al.*, 2011) found that especially the highest temperature region is critical concerning the grain growth during diffusion bonding of OFE copper samples. Initially, all the test samples had a grain size of 100 µm. After diffusion bonding for about two hours under hydrogen atmosphere, it was observed that the samples that were bonded at very high temperature of 1040 °C showed over 20 times larger grain size than initially. Moreover, their average grain size was even five times larger than of those that were bonded lower temperature of 820 °C. Optical microscopy pictures of etched samples with clearly visible difference in the grain sizes are presented in the figure 14. On the right is a sample that was bonded at high temperature and on the left is a sample that was bonded at lower temperature. (Aicheler *et al.*, 2011)

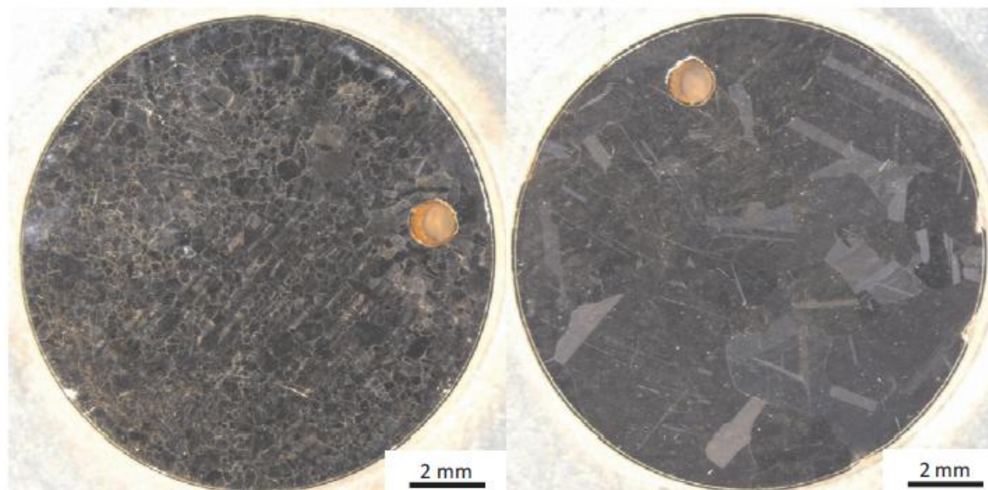

Figure 14. Optical microscopy pictures of OFE copper samples bonded at different temperatures (Aicheler *et al.*, 2011).

3.3.2 Material behaviour in diffusion bonding

When material properties are symmetric in all directions, the material behaviour is said to be isotropic. In turn, if the properties vary with different crystallographic orientations, the material behaviour is anisotropic. In FCC metals like copper, the atomic diffusion mechanisms are considered to be isotropic. Also the diffusion rate is assumed to be independent of the direction of material flow. On the contrary, some short-circuit diffusion mechanisms show anisotropic features due to the fast diffusion paths that can channel the direction of material transport in an anisotropic way. Forming of metals usually leads to a situation, where the grains are distorted and elongated in one or more directions. Consequently, forming can change the material behaviour from isotropic to anisotropic. Diffusion bonding can also be considered as one type of material forming, since high temperature, constant pressure, and long process duration are present. Some references suggest that when deformations are lower than the grain size, material behavior could be anisotropic. However, as more experimental study would be needed on this topic, isotropic material behaviour is assumed in diffusion bonding. (Butrymowicz *et al.*, 1973, NDT Resource Center, 2013, Kazakov, 1981: 54-60)

3.4 Thermo-mechanical deformations in diffusion bonding

There are three types of macro-scale deformations that are related to diffusion bonding: thermal expansion due to heating, elastic and plastic deformations due to applied stress, and creep. All of these have some influence on the final shape of the bonded components depending on the material and the bonding parameters. Considering the diffusion bonding of CLIC accelerating structure disks the applied pressure is around 0.1 MPa, while the yield strength of pure copper is 69 MPa. Thus, in this case the elastic and plastic deformations due to applied stress can be neglected. Also from thermal expansion the copper disks are completely reverted after controlled cooling. In this context it is appropriate to mention that after assembling the whole accelerating structure by diffusion bonding and brazing it is vacuum baked at 650 °C for at least 10 days to release any residual stresses. (Aicheler *et al.* 2012, ASM Handbook, 1992) So after all, the only deformation mechanism that has to be taken into account considering the unwanted permanent deformations due to diffusion bonding is creep.

4 CREEP IN DIFFUSION BONDING

Principles and basic mechanisms of diffusion were introduced in the chapter 2. It is necessary to understand the basic principles and mechanisms of diffusion to be able to study creep. In this chapter, the definition and principles of creep are introduced as well as the basic mechanisms of diffusional creep that are related to diffusion bonding.

4.1 Definition of creep

Creep is generally defined as the time-dependent deformation of materials. An increase in the applied stress or temperature accelerates the creep strain. Creep effects take place when the operating temperature of metal is about 0.3 to 0.4 of melting temperature. After that, creep rate increases exponentially with temperature. Applied stress, long timescale, and high temperature lead to vacancy formation and movement in the metal lattice. Thus, the rate of diffusion determines straightly the rate of creep. (Kraus, 1980: 1-10, Frost & Ashby, 1982, Ashby & Jones, 1980: 171-179)

4.2 Creep curve

Most elevated temperature creep tests are conducted by measuring the creep strain as a function of time under a constant load. The resulting creep strain curve can be divided into three regions: primary, secondary, and tertiary stage of creep. Creep strain curve of this general shape is presented in the figure 15. The starting point of the curve at time zero shows the initial elastic strain occurring in the structure right after applying the load. In the primary stage, the creep strain is increasing slowly over time due to changes in the substructure of the metal. The secondary stage is called the steady-state creep region. In this state the creep strain is considered to increase almost linearly as dynamic balance occurs between strain hardening and recovery processes, while the substructure of the metal remains stable. After a certain point, the metallurgical instabilities and weakening processes take control over recovery processes, and strain-rate turns to rapid growth yielding eventually to the rupture of the metal. (Hertzberg, 1976: 132-133, Ashby & Jones, 1980: 173)

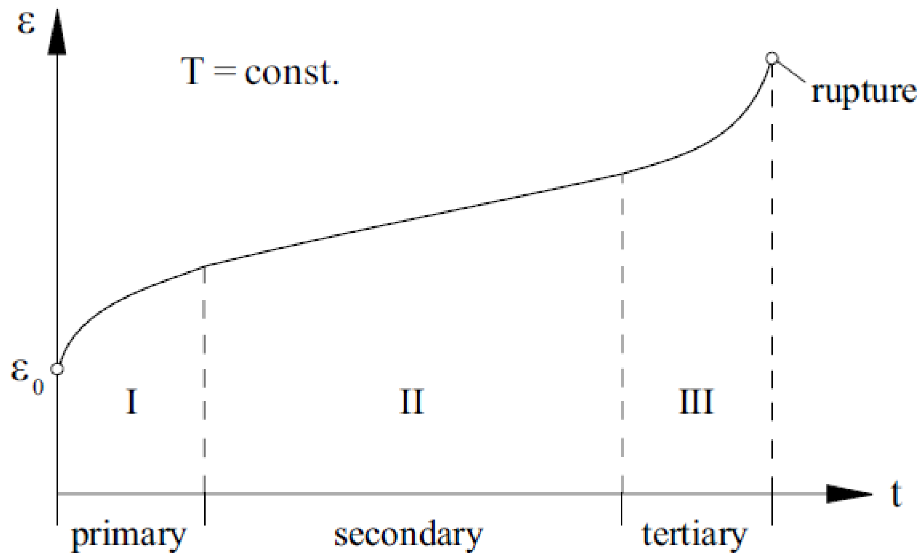

Figure 15. Creep strain as a function of time under constant load. Typical creep strain curve for metals. (Betten, 2008: 51)

4.3 Effect of stress and temperature

There are two main creep mechanisms that occur in different circumstances. *Dislocation creep* is based on plastic deformation of the crystal lattices. Atoms climb through dislocations from lattice plane to another and glide along the plane to a next climbing channel. For dislocation creep to take place in solid metals, a relatively high stress is required. The dislocation creep rate is highly dependent on applied stress and follows the so called power-law creep behaviour. (Ashby & Jones, 1980: 187-191)

Deformation mechanism map demonstrates the relation between stress and temperature, and their effect on deformation behaviour. Deformation mechanism map of pure copper is presented in the figure 16. From the map it can be seen that at high temperature and low stress deformation occurs mainly by diffusional flow of material (*diffusional creep*). The material is plastically "flowing" with a certain strain rate. Diffusional flow described earlier is driven by a gradient of free energy (chemical potential) created and increased by the applied heat and stress. (Frost & Ashby, 1982)

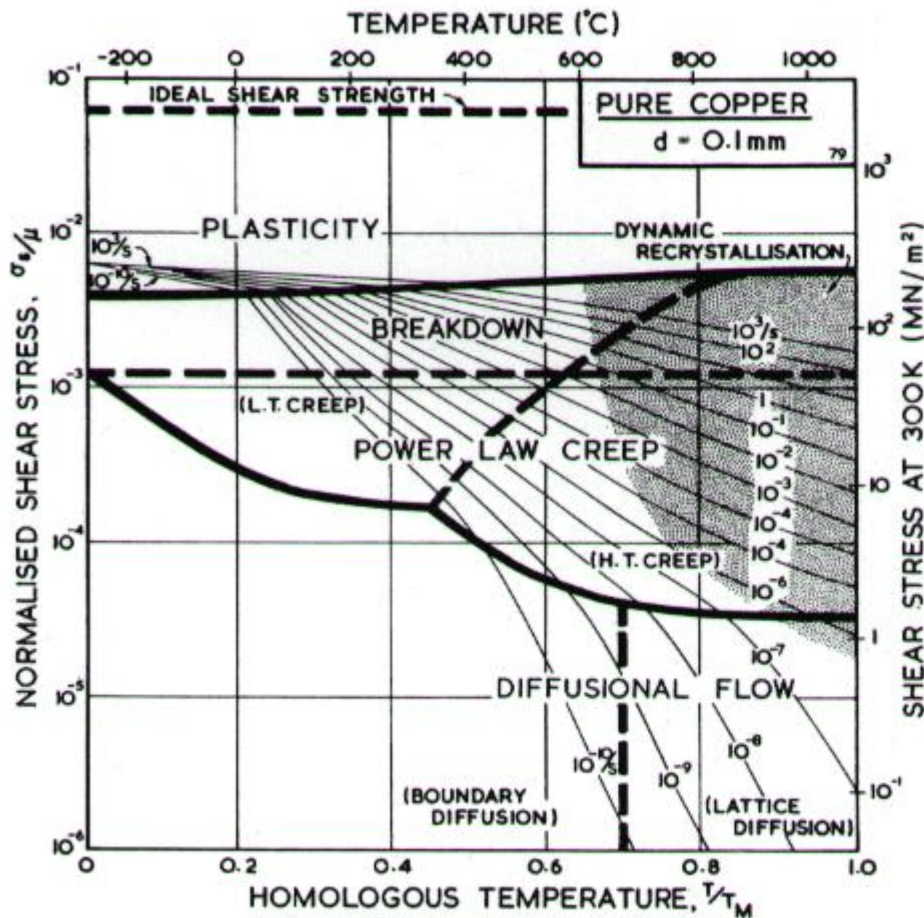

Figure 16. Deformation mechanism map for pure copper (Frost & Ashby, 1982).

Both the vacancy diffusion and the interstitial diffusion through the lattice are initially random-directional. It is assumed, that each individual atomic jump is independent of all others. Thus, the moving atom does not have any "memory" and it moves randomly to every possible directions. For more thorough information about this so called *random walk theory*, refer to Mehrer's *Diffusion in Solids* (2007). Applying stress to the random diffusional movement yields the material flow to take a direction. (Mehrer, 2007: 54-63)

Several studies suggest that diffusional creep requires a certain level of stress before taking place. In pure metals, the mobility of different kind of defects, like grain boundary dislocations, is normally high. Only at very low applied stresses, the self-stress of the boundary dislocations comes significant and limits their mobility. The energy stored in this self-stress is proposed to be the source of the *threshold stress* for diffusional creep. In other words, the applied stress has to be sufficient to overcome the

self-stress of the boundary dislocations. This threshold stress is observed to be highly temperature dependent. In pure metals, the threshold stress increases with decreasing grain size and with decreasing temperature. For example, in copper with the grain size of 35 μm, at temperature of around 550 K, creep effects are reported to activate as the applied stress is around 0.50 MPa. (Kassner, 2009: 98, Frost & Ashby, 1982: 124-126)

4.4 Mechanisms of diffusional creep

The conditions of high temperature and very low stress in diffusion bonding are favorable for diffusional creep rather than dislocation creep (Ashby & Jones, 1980: 187-191). There are two types of diffusional creep mechanisms, depending on whether the transport of material occurs along the grain boundaries or through the grains. At low temperatures, the creep strain-rate is controlled by *grain-boundary diffusion* (*Coble creep*). At higher temperatures, the vacancies flow through the grains and *lattice diffusion* controls the creep strain-rate (*Nabarro-Herring creep)*. (Herring, 1950, Coble, 1962) The paths of material flow in both types of diffusional creep are presented in the figure 17. Direct microstructural evidence for the occurrence of diffusional creep in OFHC copper is showed, for example, by McNee (*et al.*, 2000) observed using scanning electron microscopy and atomic force microscopy. In that study, creep tests of various setups were performed at high temperature and low stress.

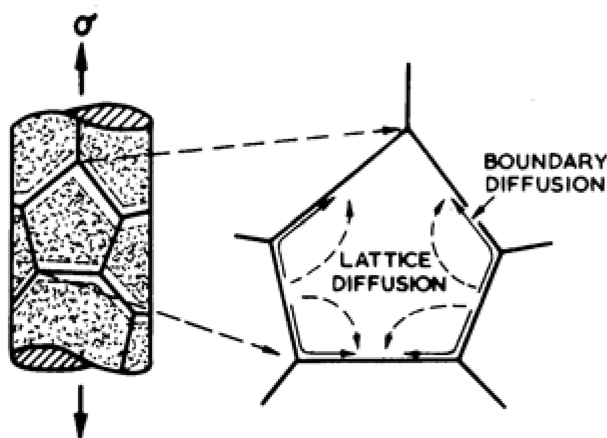

Figure 17. Paths of diffusional flow in lattice diffusion and grain boundary diffusion (Frost & Ashby, 1982: 15).

4.4.1 Coble creep formulation

At temperatures below 0.7 of the melting point, creep is controlled by diffusion of vacancies along the grain boundaries. As discussed already earlier, the grain boundaries can be connected together forming short-circuit diffusion paths. (Kassner, 2009: 86, 97) Creep strain-rate originally formulated by Coble (1962) is defined as

$$\dot{\varepsilon}_{Co} = A_{Co} * \frac{D_{gb} * \Omega}{d^3 * k * T} * \sigma \tag{1}$$

where D_{gb} is the coefficient for grain boundary diffusion, A_{Co} is the Coble's constant, Ω is the atomic volume, d is the grain diameter, k is the Boltzmann's constant, T is the absolute temperature, and σ is the stress. (Kassner, 2009: 97)

4.4.2 Nabarro-Herring creep formulation

At temperatures above the Coble creep region, vacancies flow straight through the grains. Creep strain-rate can be calculated from the equation

$$\dot{\varepsilon}_{NH} = A_{NH} * \frac{D_l * \Omega}{d^2 * k * T} * \sigma \tag{2}$$

where D_l is the coefficient for lattice diffusion. (Kassner, 2009: 97)

Diffusion coefficients for both Coble creep and Nabarro-Herring creep are defined by Fick's first law (Ashby & Jones, 1980: 183)

$$D = D_0 * e^{-Q/R*T} \tag{3}$$

where D_0 is the diffusion constant, Q is the activation energy for diffusion, and R is the universal gas constant. Both the diffusion constant and the activation energy are experimentally defined and depend on the diffusion mechanism in question. For example, in vacancy diffusion, the activation energy consists of total energy required to formation and movement of vacancies. For pure copper, the activation energy for self-diffusion is reported to be between 200 and 210 kJ/mol. (Butrymowicz *et al.*, 1973, Hertzberg, 1976: 142, Ashby & Jones, 1980: 184)

4.5 Influence of grain growth on creep

The influence of grain size on the diffusional flow is usually straightforward in pure metals: the smaller the grain size, the faster the strain-rate. Generally, creep strain-rate depends on grain size by the proportion of $(1/d)^m$, where the exponential m = 0 in case of dislocation creep, and m ≥ 2 when diffusional creep is dominant. The strain-rate is proportional to d^{-2} in the Nabarro-Herring creep model while it is proportional to d^{-3} in the Coble model. Grain size dependency of the creep strain-rate for copper is presented in the figure 18. (Wilshire & Palmer, 2001, Kassner, 2009: 97-98) Grain size affects also on the threshold stress for creep mechanisms. In pure metals, the threshold stress decreases with increasing grain size. Some references suggest that at some point, a new creep region would appear as the diffusional creep fields are suppressed by a large grain size. After that the grain size would not effect on the creep rate anymore (*Harper-Dorn creep*). This controversy creep mechanism is not fully understood and seems to involve dislocations within the grain interiors. Some authors still question the whole existence of the Harper-Dorn creep. (Frost & Ashby, 1982, Blum & Maier, 1999, Ginter & Mohamed, 2002, Kassner M. *et al.*, 2007, Kassner, 2009: 101-111)

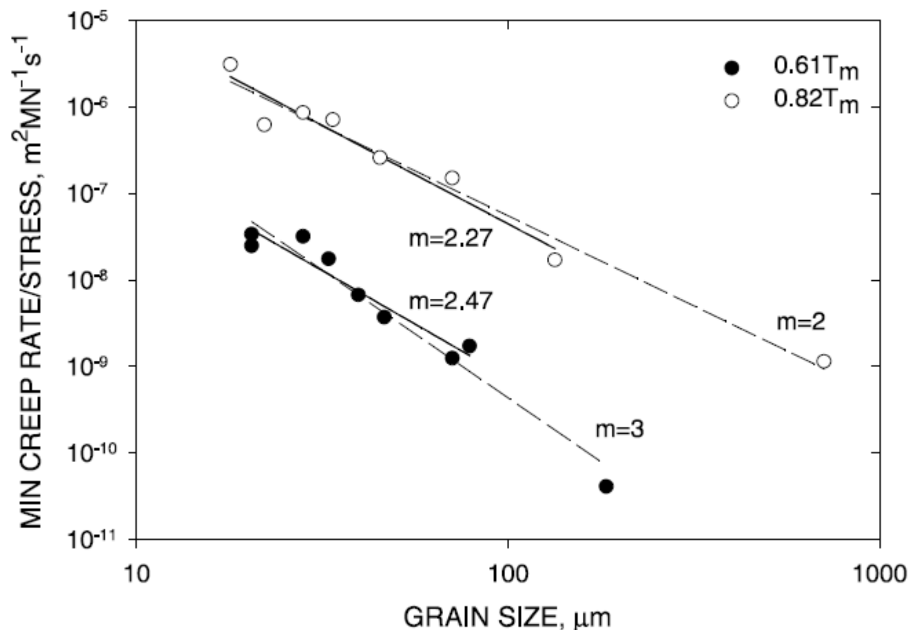

Figure 18. Grain size dependence of the creep strain-rate for copper (Wilshire & Palmer, 2001).

5 CASE STUDY

In this case study, an experimental test on diffusion bonding of the CLIC accelerator structure disks is introduced with a corresponding finite element simulation. Also a comparison between the results of the experimental test and simulation is presented. The aim of the experimental test and finite element simulation was to optimize the diffusion bonding process parameters so that the permanent deformations of the disks are reduced as low as possible, tolerances being few micrometers.

5.1 Objective

According to the current scheme, the Compact Linear Collider (CLIC) will be manufactured in modules and installed to a 50-kilometer-long tunnel underground. Each 2-meter-long module consists of the components that are needed for the function of the accelerator. Accelerating structure is one of these components. It is composed of 26 regular disks and two coupler disks of which each has an external diameter of 80 mm. Disk material is oxygen-free electronic (OFE) copper. Attached to the disk stack, there are waveguides, vacuum manifolds, flanges, cooling circuits and so on. Figure 19 illustrates the 3D view of a single CLIC accelerating structure. (Aicheler *et al*. 2012)

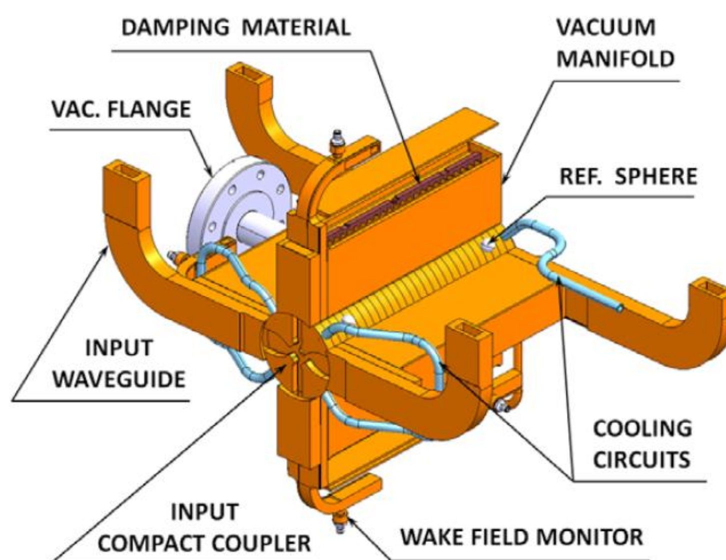

Figure 19. 3D illustration of the CLIC accelerating structure (Aicheler *et al*. 2012).

In this case study the main focus is in the disk stack at the core of the accelerating structure. The disk stack in itself is presented in the figure 20. The assembly of the stack is implemented by diffusion bonding. The total number of accelerating structures in the designed final CLIC accelerator is going to be over 142 000. Therefore, it is reasonable to study and optimize the diffusion bonding process parameters before operating the mass production. (Aicheler *et al*. 2012)

Figure 20. CLIC accelerating structure disk stack (Aicheler *et al*. 2012).

One part of the process optimization is to learn how to control the unwanted permanent deformations in the disks due to diffusion bonding. During bonding, disks undergo elastic and plastic deformations due to high temperature and low pressure between contact surfaces. At the moment, the main cause of unwanted permanent deformations is considered to be diffusional creep. In order to have adequate control on the creep deformations it is important to understand the effect of temperature and pressure on creep mechanisms. In this case study, a finite element model including the formulation of diffusional creep by Coble and Nabarro-Herring is presented. Moreover, the results of the simulation are compared with the results of recently conducted experimental test.

5.2 Experimental test

Various experimental tests on diffusion bonding of CLIC accelerating structure disks have been conducted in CERN. Recently, an experimental test was carried out in order to learn more about the macroscopic thermo-mechanical deformations of the disks, to validate the developed finite element model, and to compare the effects of different bonding pressures on the bonding joint quality. (Moilanen, 2012a) In the experimental test two different disk assemblies were used. However, in this case study, the bonding setup and results, and the finite element simulation are presented for just one of the assemblies as an example. The bonding was conducted at Bodycote in Annecy, France.

5.2.1 Test assembly

The test assembly consisted of two different accelerator structure disks: coupler disk and symmetric disk. Both of the disks have the same external diameter of 74 mm and in the centrum there is a small round hole for the particle beam, the iris. The coupler disk is thinner than the symmetric disk and it has waveguides only on the other side of it; the other side being flat. The symmetric disk has similar waveguides on both sides. A sketch of the test assembly is presented in the figure 21 below. A photograph of the test assembly is showed in the figure 22.

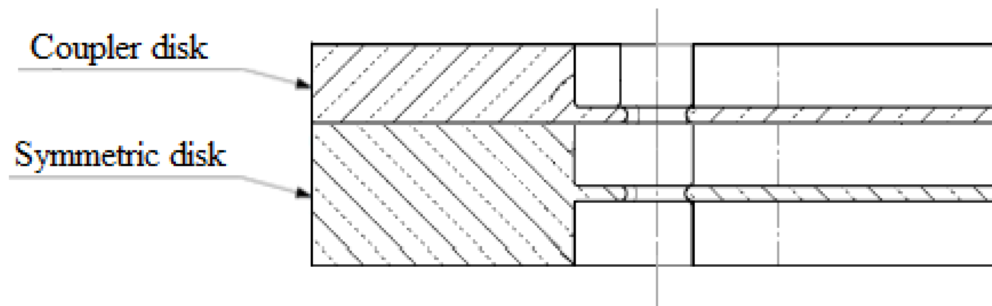

Figure 21. Sketch of the test assembly. (Courtesy of CERN)

Figure 22. Photograph of the test assembly. (courtesy of CERN)

5.2.2 Diffusion bonding parameters

Diffusion bonding was done under hydrogen atmosphere at 1025 °C temperature. Holding time was 1.5 hours. In total, the thermal cycle took about 12 hours, added the time elapsed for heating and cooling. The bonding temperature curve is presented in the figure 23. The melting temperature of pure copper is 1083 °C, thus the bonding temperature in this case is very high: about 0.95 of the melting point. (ASM Handbook, 1992)

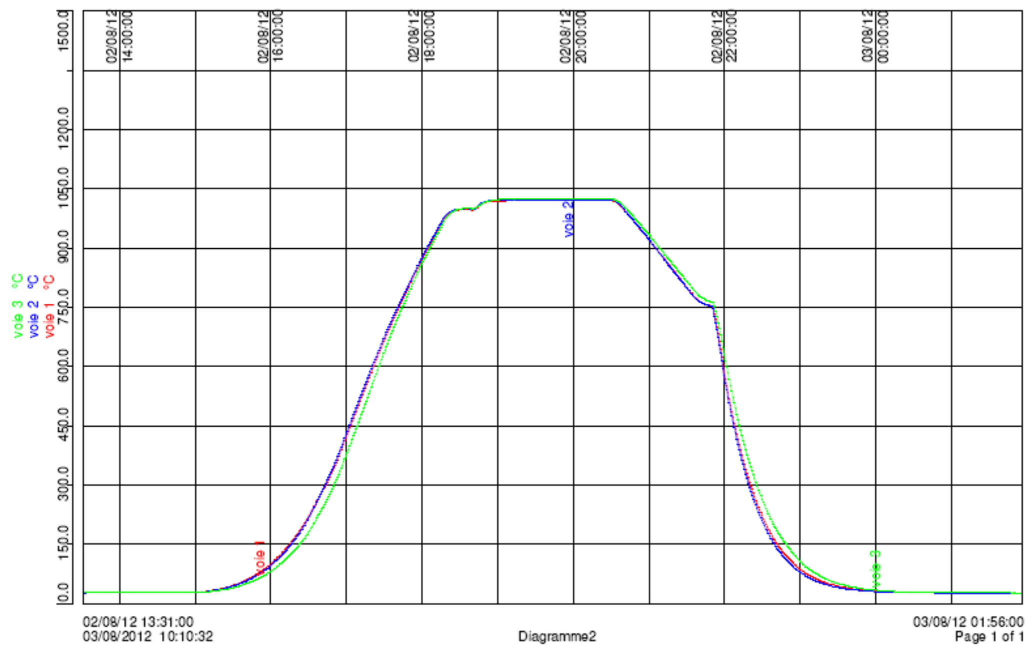

Figure 23. Bonding temperature curve. (Courtesy of CERN)

Contact area between coupler disk and symmetric disk is A = 2372 mm². Required bonding pressure was p = 0.05 MPa, so the weight to apply can be calculated by

$$F = p * A \tag{4}$$

As a result, the added weight is F = 0.05 N/mm² * 2372 mm² = 119 N. Thus, the mass load to apply is about 12.1 kg. The load was applied on top of the disk stack to produce a constant pressure between the contact surfaces. In diffusion bonding of CLIC accelerating structure disks, ceramic or graphite pads are used between the applied load

and disk stack as well as below the disk stack. The pads provide a flat and insulated base for the bonded disks and transfer the load creating a uniformly distributed bonding pressure between the disks. Stack of two disks with 12 kg applied load and graphite pads is presented in the figure 24.

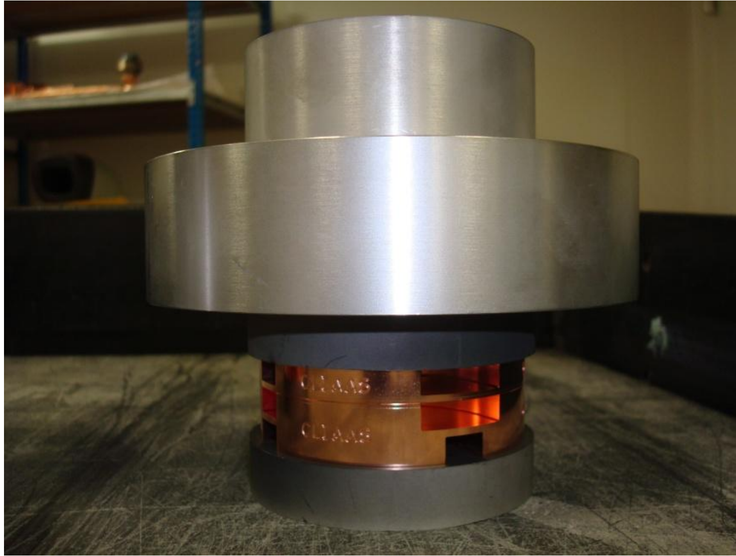

Figure 24. Stack of two copper disks, graphite pads and loads. (Courtesy of CERN)

5.2.3 Material preparation

Good surface finish is required to achieve a tight bonding joint. With rough surfaces, more pressure would be needed to achieve a sufficient contact. With current parameters, the shape tolerance of the disks is ± 2.5 μm and the surface roughness is 0.025 μm. Moreover, the accelerating structures have to be aligned to an accuracy of 5 μm. The fabrication and alignment tolerances base on the requirements of beam dynamics. Bonding joint quality is also dependent on the cleanness of the bonded surfaces. Therefore, the disks are cleaned before bonding by a dedicated cleaning procedure including chemical etching and ultrasonic cleaning in de-ionized water and in alcohol. (Aicheler *et al.*, 2012)

5.2.4 Experimental test results

Dimensional control of the disks was done before and after bonding using 2D optical microscope. Variation of the external diameter is presented in the table 1. External

diameter was measured separately from the thin and thick areas of the disks, so in the table there are two measurements for both disks. Average deformation of the external diameter was -3 µm. Similar shrinkage of the disks is observed also in previous experimental tests and it is assumed to be caused by the applied pressure combined with high temperature and long process duration. (Higo *et al*., 1996, Rossi, 2011 and 2012)

Table 1. External diameter measured before and after bonding (Moilanen, 2012d).

Disk	#	∅ [mm] Before	∅ [mm] After	Δ∅ [mm]	Δ∅ [mm] Average
Coupler	Thick	74.00	74.00	-0.001	-0.003
Coupler	Thin	74.00	74.00	-0.003	
Symmetric	Thick	74.00	74.00	-0.002	
Symmetric	Thin	74.00	73.99	-0.007	

Results of the thickness measurements are presented in the table 2. Thickness was also measured separately from the thin and thick areas of the disks. In the table, thickness before bonding is a sum of the thicknesses of the disks. After bonding, it was observed that the average deformation of thickness was -7 µm.

Table 2. Thickness measured before and after bonding (Moilanen, 2012d).

#	Area	T [mm] Before	T [mm] After	ΔT [mm]	Δ∅ [mm] Average
Assembly	Thick	23.31	23.30	-0.008	-0.007
Assembly	Thin	16.80	16.79	-0.006	

Original reports of the dimensional control can be found from (Moilanen, 2012b, 2012c). As a conclusion, it can be stated that permanent deformations of the disks due to diffusion bonding were generally low (< 10 µm) or negligible.

Bonding joint quality was analysed after the experimental test and following results were obtained. Ultrasound test and crossing grains analysis showed marks of

insufficient bonding joints in some areas. In the ultrasound scan in the figure 25, the defected areas are localized in the areas marked in red, yellow and green. In dark blue areas the contact is fine and no discontinuities were observed. (Pérez, 2012) It is possible that the flatness of the disks before bonding was not sufficient in all areas, which could have caused the observed discontinuities (Moilanen, 2012b and 2012d).

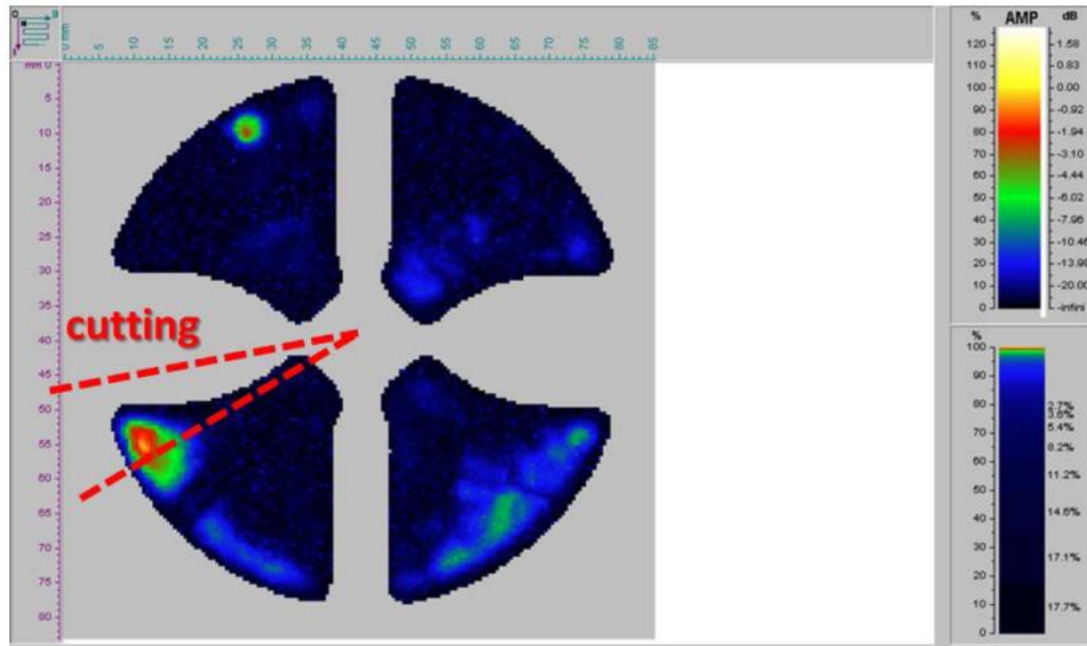

Figure 25. The results of ultrasound scan for the test assembly (Pérez, 2012).

From the pictures of crossing grain analysis (figure 26 below), it was observed that there were no crossing grains along the bonding interface. Although the non-presence of crossing grains does not necessarily mean failed bonding, with the ultrasound scan results this gives support to the idea of an inhomogeneity of the bonding. (Pérez, 2012)

Figure 26. Crossing grain analysis picture (Pérez, 2012).

5.3 Finite element simulation

Aim of the finite element simulation was to model the macroscopic thermo-mechanical deformations of the disks due to the bonding process. With a valid simulation it would be possible to predict the deformations and test different bonding parameters and assemblies without having to conduct time-consuming and expensive experimental tests in every step. The finite element simulation of diffusion bonding process included nonlinear material model, time and temperature dependent variables, contacts and large temperature variation combined with long process duration. The modelling was done using ANSYS 14.

5.3.1 Material model

First of all, there was no suitable material in the ANSYS material library. Instead, a Copper Alloy Non-Linear was selected as a base for the OFE copper and the required material properties were defined manually as functions of temperature. Material properties for OFE copper can be found from ASM Handbook (1992). In the lack of experimental data about the material behaviour in diffusion bonding, isotropic material behavior was assumed in the present finite element analysis. Creep was included in the model by defining the diffusional creep strain-rate equations of Coble and Nabarro-Herring. These two were the only creep models included in the current model, since there are uncertainties about the existence of the Harper-Dorn creep. Temperature-dependent diffusion coefficient values for the creep equations were calculated using Wolfram Mathematica software and imported to Ansys as tabular data.

5.3.2 Geometry

Modelled test assembly was the same as in the experimental test. Models of the accelerating structure disks are presented in the figure 27. Real 3D geometries of the disks were imported to ANSYS from Catia V5 software. Small features as chamfers were suppressed in ANSYS 14 Design Modeller using face delete. Since the test assembly is axisymmetric, only one quarter of the stack was modelled. Simplified geometry for the simulation is presented in the figure 28.

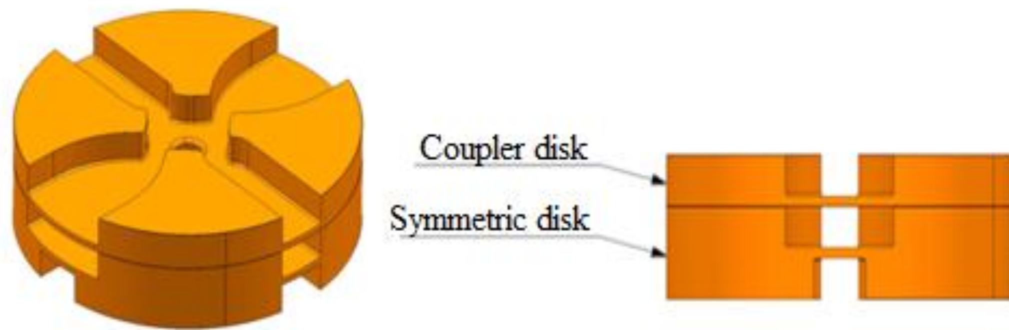

Figure 27. 3D models of the accelerating structure disks provided by Anastasiya Solodko (CERN).

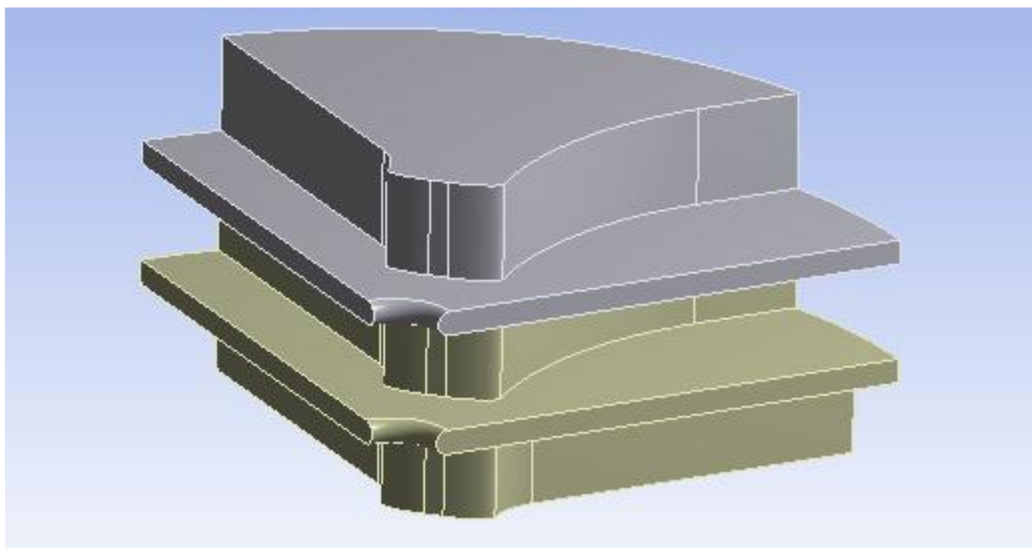

Figure 28. Simplified geometry for the simulation (Moilanen, 2012d).

5.3.3 Load and boundary conditions

Loads and boundary conditions for the simulation are presented in the figure 29. Constant pressure load was set on top of the disk stack [A]. Symmetry conditions were defined on the cut surfaces [B] and [C]. Frictionless support was set on the bottom of the stack [D], and frictional contacts with friction coefficient of 1.0 were set between the disks.

Temperature profile measured during bonding tests was imported to the finite element model and set as thermal condition to all bodies. To get comparable results, same environment temperature was defined for the first and last load step.

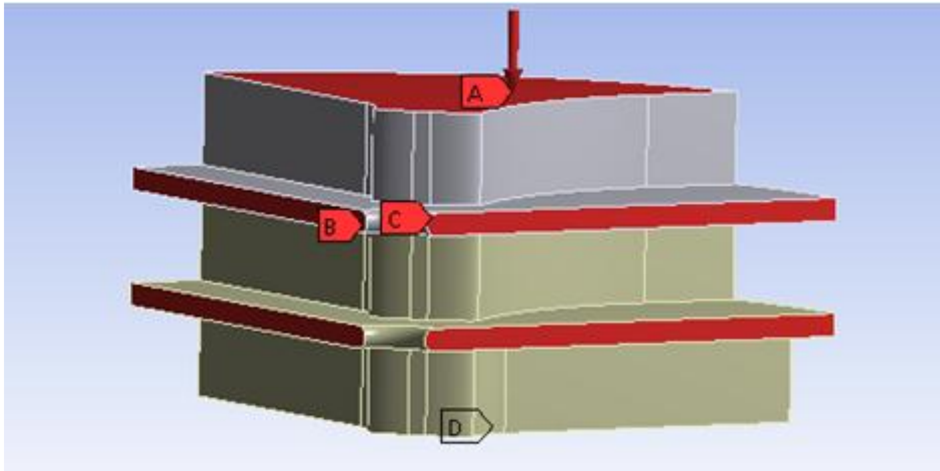

Figure 29. Loads and boundary conditions (Moilanen, 2012d).

5.3.4 Mesh

Finite element mesh was generated using quadratic tetrahedron elements with size defined as 1.5 mm. Mesh in the figure 30 contains over 43000 elements and 65000 nodes. Mesh convergence study was performed in order to obtain an accurate solution without overly increasing the computational time.

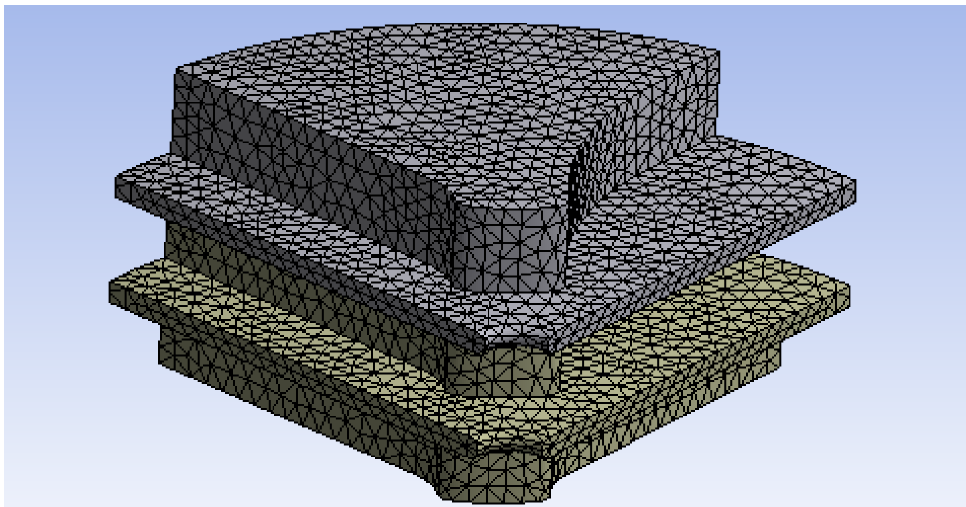

Figure 30. Finite element mesh (Moilanen, 2012d).

5.3.5 Simulation results

Results of the finite element simulation are presented next. Deformation of radius is showed in the figure 31. Deformation increases radially and is distributed rather symmetrically around the circular outer shape of the stack. Maximum radial deformation was slightly over 6 µm on the outer surface of the coupler disk. Thus, the deformation of external diameter in total was 13 µm.

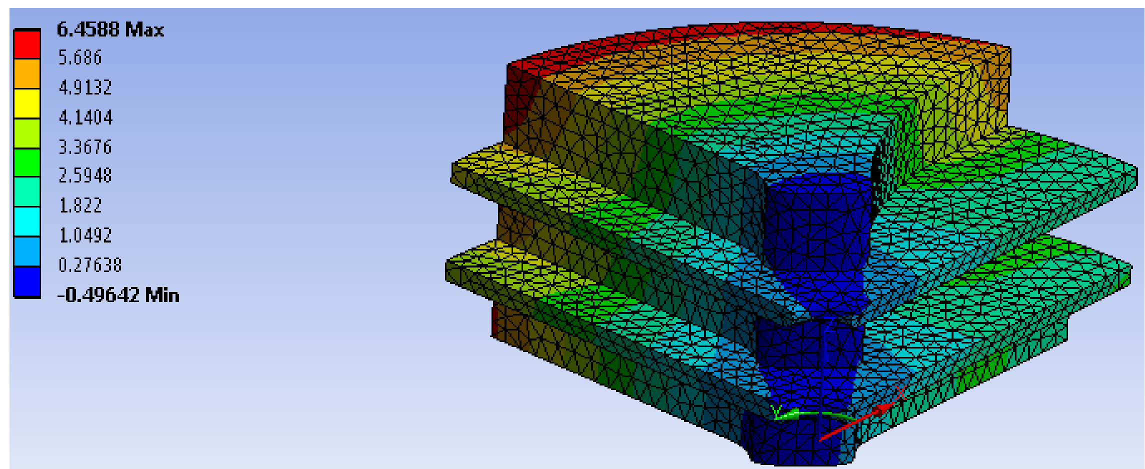

Figure 31. Radial deformation in µm (Moilanen 2012d).

Deformation of thickness is showed in the figure 32. Maximum deformation of thickness was about -11 µm on the top of the disk stack. There was also a minor positive deformation of 2 µm in one of the waveguides of the symmetric disk.

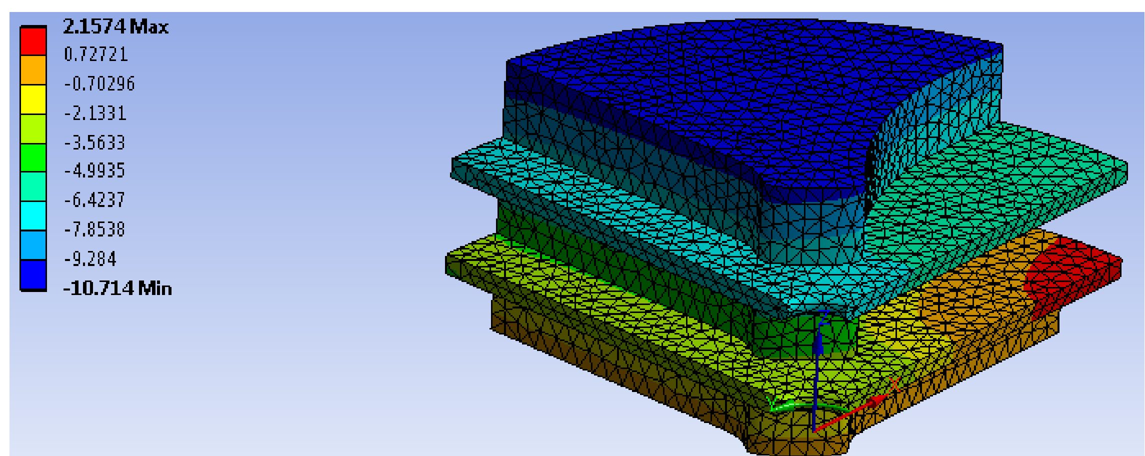

Figure 32. Deformation of thickness in µm (Moilanen 2012d).

5.4 Validation

Finite element simulation showed some permanent deformations in external diameter and thickness of the bonded disk stack. Meanwhile, in the experimental tests the deformations were very low or even negligible. Comparison of the results of experimental test and simulation are presented in the table 3.

Table 3. Comparison of the results of experimental test and simulation.

	ΔØ [mm]	Δ Thickness [mm]
Experimental test	-0.003	-0.007
FEM simulation	+0.013	-0.011

Some of the deformations that were seen in the simulation could be explained by inaccuracy due to the numerical solution techniques used by the software. Still, especially the deformation of external diameter is quite significant comparing with the test result. Hence, in this case another explanation for the deformations is needed.

As discussed already in the chapter 4, several references suggest there is a certain threshold for stress, below which creep mechanisms do not activate. The value of the threshold depends on material properties, temperature and so on, and thus it cannot to be defined generally. At the moment, there are two experimental tests related to the bonding of CLIC accelerator disks in which a low applied stress of 0.05 MPa was used and no deformations after bonding were observed. Instead, deformations have been observed in previous bonding tests using a pressure of 0.10 MPa or higher. Some of these results have been documented in CERN EDMS documents 1165715 and 1176125.

This gives a suggestion for threshold stress that would be around 0.05 MPa in the present diffusion bonding case. In turn, in the current finite element simulation the creep strain rates are calculated even for the lowest stresses, which could give a reasonable explanation for the deformations seen in the finite element simulation.

6 CONCLUSIONS

Various microstructural changes in metals are based on the diffusion phenomena. The most important diffusion mechanism in pure metals is the vacancy mechanism. Other diffusion mechanisms are, for example, interstitial and short circuit mechanisms. Diffusion phenomena is also strongly present in diffusion bonding, an advanced solid-state joining process in which the surfaces of two components are brought to contact with each other and heated under a pressing load in a controlled environment. During the process, contact surfaces are bonded by atomic diffusion across the interface and one solid piece is formed.

The conditions of high temperature and low stress in diffusion bonding are favorable for creep effects to take place in the bonded metals. An increase in the applied stress or temperature accelerates the creep strain. The most important creep mechanisms in diffusion bonding are the Coble creep operating at temperatures below 0.7 of melting temperature, and the Nabarro-Herring creep dominating at higher temperatures. Both of these diffusional creep mechanisms are highly dependent on the grain size of metal. Observations of an excessive grain growth during diffusion bonding have given a rise to the idea of alternate creep theories that would be independent of the grain size. One candidate for this kind of creep model would be the controversial Harper-Dorn creep. Nevertheless, the effect of excessive grain growth on creep needs further investigation.

In the case study, a finite element simulation of diffusion bonding process was introduced. Also an experimental test conducted in CERN Compact Linear Collider - project was presented. In the tests, accelerator structure disks made of OFE copper were joined together by diffusion bonding at high temperature and very low stress. However, in the inspection of the bonding joint, some defected areas were found and it was stated that the joint may be insufficient.

The permanent deformations that were observed in disks external diameter and thickness after bonding were compared with the results of the finite element simulation. As a conclusion, a slight inconsistency between the results was noted. Simulation showed some deformations while in the experimental test the observed deformations

were very low or negligible. Therefore, the results of the recent experimental test are not useful to validate the creep model used in the finite element analysis. In turn, these results are promising to confirm the perception that there is a threshold stress for creep and that the threshold value would be around 0.05 MPa in diffusion bonding of CLIC accelerating structure disks.

In future bonding tests, it would be reasonable to test the effect of just a slightly higher bonding pressure. Also improving the surface flatness of the disks should be taken into consideration. After all, the success of the bonding joint is highly dependent on the quality of the bonded surfaces. Especially when using very low bonding pressure the importance of the surface flatness is highlighted as it directly affects the state of the initial contact before any surface asperities are deformed by the applied stress and heating. Therefore, the starting value of the flatness is imposing the bonding pressure to be used and thus greatly affects the permanent deformations in the disks.

The simulation and the experimental tests together suggest that with a subtle process optimisation it could be possible to achieve low deformations *and* a sufficient bonding joint at the same time. Also bonding pressure of 0.05 MPa should be tested again with improved surface flatness of the disks, or with different test assemblies. After all, the observed deformations were promisingly low, and even though the bonding joint was partially deficient, it was not completely failed.

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APPENDIX

TIIVISTELMÄ APPENDIX 1

Tämä kandidaatintyö liittyy Euroopan hiukkasfysiikan tutkimuskeskuksen (CERN) lineaarikiihdytinprojektiin (CLIC), jonka tavoitteena on suunnitella ja rakentaa seuraavan sukupolven lineaarinen hiukkaskiihdytin. Suunnitteilla olevan lineaarisen hiukkaskiihdyttimen keskeisimpiin osiin kuuluvien kiihdytinrakenteiden kokoonpano toteutetaan pääosin diffuusioliittämällä ja -juottamalla. Kiihdytinrakenteet altistuvat hiukkaskiihdyttimen käytössä erittäin voimakkaille sähkö- ja magneettikentille. Rakennemateriaalina käytetäänkin hapetonta kuparia sen hyvän sähkön- ja lämmönjohtavuuden ansiosta. Diffuusioliittämisen seurauksena liitettäviin kupariosiin voi jäädä ei-toivottuja pysyviä muodonmuutoksia, joiden minimoimiseksi metallin käyttäytyminen diffuusioliittämisen aikana olisi tunnettava mahdollisimman tarkkaan. Tässä kandidaatintyössä käsitellään hapettoman kuparin diffuusioliittämisessä esiintyviä virumisilmiöitä.

Kiinteässä metallissa atomit ovat pakkautuneet yhteen muodostaen kolmiulotteisia kiderakenteita. Kiderakenteiden nimet kuvaavat atomien keskinäistä sijaintia rakenteessa. Yleisimmät kiderakenteet ovat pintakeskeinen kuutiollinen rakenne, tilakeskeinen kuutiollinen rakenne sekä heksagonaalinen tiivispakkausrakenne. Kiinteällä kuparilla on huoneenlämpötilassa pintakeskeinen kuutiollinen rakenne.

Diffuusiolla tarkoitetaan atomien tai partikkelien liikettä ympäröivässä materiaalissa. Monet metallien mikrorakenteen muutokset perustuvat fysikaalisesti diffuusioilmiöön. Kiinteässä metallissa diffuusio perustuu metallin kiderakenteen virheisiin eli hilavirheisiin. Hilavirheet voidaan jakaa pääsääntöisesti pistemäisiin ja viivamaisiin virheisiin. Erilaisia pistemäisiä hilavirheitä ovat muun muassa vakanssit eli kidehilan aukot ja välisija-atomit. Viivamaisia virheitä ovat särmä- ja ruuvidislokaatiot. Muita hilavirheitä ovat esimerkiksi raerajat ja vapaat pinnat. Puhtaassa metallissa yleisimpiä hilavirheitä ovat vakanssit. Sen sijaan seosmetallien ja epäpuhtauksien yhteydessä välisija-atomit ovat hyvin yleisiä hilavirheitä.

Puhtaissa metalleissa tapahtuvaa diffuusiota kutsutaan itseisdiffuusioksi. Metalleissa esiintyvät diffuusiomekanismit liittyvät läheisesti hilavirheisiin. Yleisimmät diffuusiomekanismit ovat välisijamekanismi ja vakanssimekanismi. Puhtaissa metalleissa vakanssimekanismi on hallitseva diffuusiomekanismi, kun taas pienten seosatomien ja epäpuhtauksien yhteydessä välisijamekanismi on yleisin. Välisijamekanismissa diffundoituva atomi käyttää kiderakenteen välisijoja etenemiseen. Vakanssimekanismissa taas kiderakenteessa oleva tyhjä atomipaikka eli aukko vaihtaa paikkaa viereisen atomin kanssa, ja prosessin toistuessa muodostuu materiaalivirtaus vastakkaiseen suuntaan, kuin mihin aukko liikkuu. Metalleissa voi esiintyä myös dislokaatioiden ja muiden rakenteellisten virheiden muodostamia nopeita diffuusioreittejä. Esimerkiksi raerajadiffuusiossa atomit liikkuvat raerajoja pitkin.

Diffuusioliittäminen on eräs diffuusioilmiön teknologinen sovellus. Diffuusioliittäminen on pitkälle kehittynyt kiinteän olomuodon liittämismenetelmä, jossa kahden metallikappaleen pinnat tuodaan lähelle toisiaan ja kappaleita kuumennetaan puristavan voiman alaisena kontrolloidussa ympäristössä. Prosessin aikana metallikappaleet liittyvät yhteen rajapinnalla tapahtuvan diffuusion vaikutuksesta. Diffuusioliittämistä voidaan käyttää samanlaisten ja erilaisten metallien sekä myös seosmetallien ja keraamien liittämiseen. Liitoksen onnistuminen riippuu pääasiassa kolmesta säädettävästä parametrista: lämpötilasta, puristusjännityksestä ja pitoajasta. Liitoksen laatu riippuu myös muun muassa materiaaliominaisuuksista, liitettävien pintojen esivalmistelusta sekä ympäristöstä, jossa liittäminen tapahtuu. Diffuusioliittämisessä tapahtuva suhteellisen pitkäkestoinen altistus korkealle lämpötilalle sekä pieni puristusjännitys aiheuttavat metalleissa virumista, joka taas saa aikaan ei-toivottuja pysyviä muodonmuutoksia yhteenliitettävissä kappaleissa. Korkean lämpötilan ja pitkän pitoajan seurauksena myös metallien raekoko voi kasvaa liittämisen aikana huomattavasti.

Viruminen tarkoittaa yleisesti ajasta ja lämpötilasta riippuvaa pysyvää viskoplastista muodonmuutosta. Virumismekanismit voidaan karkeasti jakaa dislokaatioliikkeen ja diffuusion aiheuttamaan virumiseen. Puhtaan kuparin viruminen korkeassa lämpötilassa ja pienen jännityksen alaisena johtuu diffuusion aiheuttamasta materiaalivirtauksesta. Viruminen kiihtyy huomattavasti lämpötilan ja jännityksen kasvaessa. Myös raekoko

vaikuttaa virumisnopeuteen: mitä pienempi raekoko, sitä suurempi virumisnopeus. Useiden tutkimusten mukaan jännitykselle on olemassa kynnysarvo, jonka alapuolella diffuusion aiheuttamaa virumista ei tapahdu. Tämä kynnysarvo riippuu muun muassa materiaaliominaisuuksista ja lämpötilasta, minkä vuoksi se voidaan määrittää ainoastaan tapauskohtaisesti. Matalissa lämpötiloissa (alle 70 % sulamislämpötilasta) virumista hallitsee raerajadiffuusio. Raerajadiffuusiossa atomit liikkuvat metallikiteiden reunoja eli raerajoja pitkin. Korkeammissa lämpötiloissa (yli 70 % sulamislämpötilasta) vakanssit virtaavat hilarakenteiden lävitse, eli vakanssidiffuusio aiheuttaa metallin virumisen.

Tapaustutkimuksessa esitellään kokeellinen tutkimus sekä vastaava elementtimenetelmäsimulaatio. Kokeellisessa tutkimuksessa kaksi kuparikiekkoa liitettiin yhteen diffuusioliittämisellä. Liittäminen tehtiin korkeassa lämpötilassa hyvin pienen kuormituksen alaisena ja pelkistävissä olosuhteissa. Lämpötila oli 95 % hapettoman kuparin sulamislämpötilasta ja painekuormitus 0.05 MPa. Diffuusioliittämiseen kuluva aika oli yhteensä yli 12 tuntia, josta aktiivista eli korkeimman lämpötilan pitoaikaa oli noin 2 tuntia. Kuparikiekoille tehtiin kontrollimittaus ennen liittämistä ja liittämisen jälkeen. Kontrollissa mitattiin kiekkojen paksuus, ulkohalkaisija, sisähalkaisija sekä pinnan tasaisuus. Diffuusioliittämisen jälkeen kiekkojen dimensioissa ei havaittu merkittäviä pysyviä muodonmuutoksia. Suurimmat muodonmuutokset olivat 8 µm luokkaa. Erityisesti ulkohalkaisijan muodonmuutokset olivat lähellä nollaa. Diffuusioliitoksen laatua tutkittaessa havaittiin, että liitos on joiltakin osin puutteellinen.

Kokeellisen tutkimuksen jälkeen esitellään elementtimenetelmäsimulaatio vastaavasta prosessista. Toimivan tietokonesimulaation avulla on mahdollista säätää liittämisprosessin parametreja ilman, että joudutaan jokaisessa vaiheessa toteuttamaan kalliita ja aikaavieviä kokeellisia tutkimuksia. Simulaatiolla voidaan myös helposti kokeilla erilaisia kokoonpanoja. Elementtimenetelmäsimulaatiota varten määriteltiin epälineaarinen materiaalimalli hapettomalle kuparille. Tämän jälkeen testikokoonpanoa vastaavat geometriat tuotiin elementtimenetelmäohjelmistoon ja niille määriteltiin tarvittavat reunaehdot ja kuormitukset. Simulaation tuloksena havaittiin, että kuparikiekkoihin oli tullut jonkin verran pysyviä muodonmuutoksia, jotka olivat

suurempia kuin kokeellisessa tutkimuksessa havaitut. Eräs mahdollinen selitys simulaatiossa nähdyille muodonmuutoksille olisi se, että virumiselle on olemassa tietty kynnysarvo. Diffuusioliittämisessä käytetty paine oli hyvin matala 0.05 MPa. Elementtimenetelmäsimulaatiossa virumisvenymät lasketaan myös kaikkein matalimmille jännityksen arvoille, mikä voisi antaa järkevän selityksen muodonmuutoksille.

Tulevissa diffuusioliittämiskokeissa olisi hyvä testata hieman korkeamman paineen vaikutusta liitoksen laatuun ja pysyviin muodonmuutoksiin. Myös kuparikiekkojen pinnan tasaisutta tulisi parantaa, koska liitoksen vioittuneet alueet voivat johtua huonosta pinnan tasaisuudesta. Kaiken kaikkiaan kokeellisessa tutkimuksessa havaitut muodonmuutokset olivat hyvin pieniä, ja vaikka liitos olikin osittain puutteellinen, se ei ollut kokonaan epäonnistunut.