MATERIAL STUDIES IN THE FRAME OF CLIC ACCELERATING STRUCTURES PRODUCTION CONDUCTED WITHIN THE MECHANICS PROGRAM TOGETHER WITH METSO OY

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Abstract

MeChanICs (Marie Curie Linking Industry to CERN) is an Industry to Academia Partnership and Pathways (IAPP) platform for precision manufacturing knowledge exchange bringing together five Finnish manufacturing companies with Helsinki Institute of Physics (HIP) and CERN. The scientific objective of MeChanICs project is to contribute to the manufacturing RTD of CLIC enabling technologies. The focus is on the design, materials, machining, brazing and assembly of a CLIC accelerating structure.

This study deals with the materials work package of the program and wants to explore the following items: 1) producing copper accelerating structures for CLIC from raw copper powder by near net shape hot isostatic pressing (HIP). 2) The feasibility to use HIP diffusion bonding of the accelerator structures as a function of surface quality and applied temperature and pressure. 3) Brazing for CLIC AS auxiliary systems, like water cooling or damping manifolds, to the disc stack by coating one of the brazing partners with an enabling layer prior to the heat treatment. The chosen coating method is electrodeposition of pure Cu/Au layers.
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1. General introduction

The Compact Linear Collider (CLIC) is an electron-positron collider for energies of 1.5 TeV - 3 TeV, currently under study at the European Organization for Nuclear Research (CERN) in Geneva [1]. It is considered as a possible successor for the Large Hadron Collider (LHC). Its task will be to conduct experiments to validate and improve precision of LHC data by colliding elementary electrons and positrons. The planned accelerating gradient of 100 MV/m will be the highest ever achieved in non-superconducting accelerating cavities [2]. This extremely high gradient is required to minimise the costs for expensive underground civil engineering for the accelerator construction [1]. The accelerating fields for the particles are induced in the Accelerating Structures (AS) by high-intensity pulsed Radio Frequency (RF) waves. Materials used for the AS are subject to pulsed surface heating arising from the electrical currents induced by the RF fields. Since CLIC will operate at room temperature, good electrical conductivity of the AS material is of paramount importance [2]. Pure electrical copper, an excellent, commercial electrical conductor has therefore been chosen as the baseline material for the AS. The drawback of this type of pure copper, however, is its low mechanical strength.

The main parts of the current design of the accelerating structure are built from high accuracy machined copper components (see Figure 1). The individual components are then joined by diffusion bonding, brazing and welding. There are competing designs – for example the accelerating structure can be assembled from discs or longitudinal quadrants/halves, as it can be seen in Figure 2. Also, experiments have been done to incorporate refractory (Mo, W) metal iris (the beam cavity in the middle of the disc) made by various methods (HIP, explosion bonding, etc...).
In materials and production point of view, the production method has a strong influence on performance feasibility. The main issue in copper parts is the oxygen content since bonding of the components is done on hydrogen atmosphere. In the case that copper material have oxides in the structure, there can happen a reaction between H\(_2\) and CuO resulting generation of water vapour inside the copper structure. This reduces the mechanical properties of copper.

Another issue are the resulting mechanical properties of the copper after the temperature and pressure cycles the material is subjected to during the assembly process, as seen in Figure 3. High mechanical properties are needed for improved fatigue resistance due to radio frequency magnetic fields that generate stresses on the copper structure. Also the surface quality at the iris is very important and needs to be machined before the bonding. The interest lies on how the different temperature cycles and gases affect the surface quality.

The main parts of CLIC components are made by copper and they need to be joined to other copper components or to other materials. Some of these joints are produced by brazing. For joining copper, the brazing material of choice is gold-copper alloy with composition of 50/50 wt.%. The brazing alloy is usually put in place in form of thin sheet of pure metal or an alloy. In case of complex forms foils
become impractical and other methods are needed. One such method is to deposit the brazing material to one or both joints by coating.

2. Motivation

Classical hot isostatic pressing (HIP) is used for powder compaction for fabrication of particular alloys, which are not producible in any other metallurgical way. This study wants to explore the possibility to produce copper accelerating structures for CLIC from raw copper powder. For this purpose, the properties of the compacted copper, as well as, the feasibility of near net shape HIPing for the accelerator structure need to be assessed.

HIP diffusion bonding is often used for of two solids in cases where the pressure is hard to arrange in conventional means. Since the accelerator structures in case are very surface quality sensitive, the effect of isostatic pressure on surfaces need to be studied in detail and will extend further a study of resulting surface quality after heat treatment (see Ref. [3]). As a logical follow-up the effect of pressure and prior surface quality on the bonding quality is of particular interest and needs further attention. Additionally to the surface quality, the resulting mechanical and microstructural properties of OFC-copper after the HIP treatment need to be systematically explored.

Brazing is often used for assembly of non-equal materials or complex. For the CLIC AS, brazing is the current baseline procedure for joining the auxiliary systems, like water cooling or damping manifolds, to the disc stack. Classical wire in groove deposition of the brazing alloy before the heat treatment is not so trivial because of the complex geometries. Coating one of the brazing partners with an enabling layer prior to the heat treatment is proposed as an alternative procedure. The chosen coating method is electrodeposition due to its relatively high deposition rate. However, due to its nature, this method is usually used for deposit pure metals. For brazing copper, one needs an alloy with melting point lower than the copper components so an alloy of Cu and Au is currently used. A diffusion treatment of pure Cu/Au layers electrodeposited before the actual brazing step could act as an in-situ formation of the desired brazing alloy. A nickel layer is proposed in order to act as a diffusion barrier for reducing the unwanted diffusion of the brazing alloy deeper to the component. The focus of this study lays on the three following issues:

- Study the diffusion behaviour of electrodeposited Au-Cu layer systems
- Study the effect of Ni-layer acting as a diffusion barrier
- Study the quality of the resulting brazing joint

3. Experimental

HIP-treatments were performed in a small scale HIP-unit at VVT, Espoo, Finland. The HIP-chamber size is Ø130 x 300 mm with maximum temperature 2000°C and 207 MPa pressure. The images of the unit and the chamber are shown in Figure 4. The treatment plan of samples consisted of five HIP-cycles with the parameters and involved samples shown in Table 1.
Table 1 Hip cycles and samples conducted for experiments.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>HIP 1</th>
<th>HIP 2</th>
<th>HIP 3</th>
<th>HIP 4</th>
<th>HIP 5</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cycles</td>
<td>900°C, 3h, 1000 bar</td>
<td>900°C, 3h, 250 bar</td>
<td>900°C, 3h, 100 bar</td>
<td>900°C, 3h, 1000 bar</td>
<td>900°C, 3h, 1000 bar</td>
</tr>
<tr>
<td>DB stack, 0.2 µm surface quality</td>
<td>-</td>
<td>X</td>
<td>X</td>
<td>X</td>
<td>-</td>
</tr>
<tr>
<td>DB stack, 0.8 µm surface quality</td>
<td>X</td>
<td>X</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>DB stack, 0.1 µm surface quality</td>
<td>-</td>
<td>X</td>
<td>X</td>
<td>X</td>
<td>-</td>
</tr>
<tr>
<td>Raw disc, Ø95x25 mm</td>
<td>X</td>
<td>X</td>
<td>X</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Diamond turned disc, Ø73x8 mm</td>
<td>X</td>
<td>-</td>
<td>X</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Diamond turned oval, Ø43/32x2 mm</td>
<td>X</td>
<td>-</td>
<td>X</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Stamp disc / Cu-powder (Figure 25)</td>
<td>X</td>
<td>-</td>
<td>-</td>
<td>X</td>
<td>-</td>
</tr>
<tr>
<td>Large steel capsule Ø114x200 mm</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>X</td>
</tr>
</tbody>
</table>

Diffusion bonding in CERN is normally done with copper components having very fine surface roughness in the order of 0.02 µm Rₐ. The smooth surface and the according flatness tolerances required are very machining intensive and hence expensive. HIP-bonding is considered as a possible alternative in order to reduce these requirements. Therefore, three sets of capsules consisting of two cover discs and a doughnut disc were prepared with 0.8, 0.2 and 0.1 µm Rₐ surface finishing. A cross section of the capsule set-up is presented in Figure 5. The capsule was designed so that there is a thick jacket and thin cover plates. The reason for this was to direct the isostatic pressure such a way that the joints are receiving axial pressure but the radial force would be as small as possible. Another point of interest in this study was the inner corner of the junction as seen in Figure 5. This area is critical for RF-performance and there are machining and assembly issues that require further development.

Figure 5 HIP-diffusion bonding capsulation of the copper stacks.
The copper discs and samples were cleaned, pickled and rinsed according to the CERN instructions [4]. The copper disc stacks were placed in AISI 316L steel cylinder and cover plates were welded gas tightly, together with low carbon steel evacuation tube. The capsules were evacuated with the help of a high vacuum turbo pump system down to a vacuum level of 3.0x10⁻⁵ mbar. The evacuation tube was then sealed by squeezing it flat with a hydraulic tool and then cut and welded with an acetylene torch. A sketch of the general arrangement (with minor modifications on each cycle) of the HIP-charge in the furnace is shown in Figure 6 whereas the process from the raw parts to the final arrangement in the furnace can be seen in Figure 7. The upper side of the bare samples was open to HIP chamber, except the diamond turned samples were placed on top of the raw disc. The heating and pressurizing was done in one hour and the cooling was free in the HIP-furnace.

![Figure 6 Study 2-HIP 2: Charging plan in the HIP-vessel. Unlike in the plan, the uppermost capsule was also standing with the evacuation tube upwards. Third carbon steel ring (not in the plan) was used between the capsule and Raw Disc.](image)

After the treatments, the HIP capsules and bare copper samples were inspected visually. The surface of the capsule was normal with green-grey appearance of the AISI 316L parts, and light grey of the carbon steel tube. The surfaces of the bare copper samples were not oxidized and the diamond turned samples were still shiny although some dirt spots could be observed on some of the surfaces. Also, the alumina plates had left their marks on the surface wherever they were in touch of the copper surfaces (see Figure 8). The centre parts of the bottom and cover of the capsules were deformed inwards (bottom about 6–7mm and cover about 4 – 5mm). The main measures of the capsules were measured with slide gauge and the outer diameter of all the capsules remained at nominal diameter of 100.0mm.
Figure 7 Copper samples for HIP 2: ultrasound cleaning of capsules in acetone, dipping of the Raw Disc in to detergent NGL 17.40 for ultrasound cleaning, pickling of the raw disc in the 50% HCl (37%) and charging of the HIP-vessel.

Figure 8 HIPped capsules and bare copper samples after the treatment.
4. Results and Discussion

4.1. The effect of HIP-cycles to solid copper properties

The high temperature of hot isostatic pressing affects copper mainly due to grain growth and by removal of the effect of cold working that is mainly softening. Since copper stays at one phase, the changes in properties are caused by grain growth. In case of high surface finishing, there is a known but not well understood effect on surface quality when the grain boundaries are affected and some faceting is often found. These phenomena must be considered in case of a component with super fine finishing needs to be treated in high temperature (for joining for instance) [5].

4.1.1. The effect on tensile strength and hardness

CERN requirements for mechanical properties of copper raw material and the properties of HIP-treated solid copper (from CERN stock) are shown in Table 2. Error! Reference source not found. shows one of the stress-extension graphs of HIP-treated solid samples with 50 mm gage length. The results are as expected considering the effect of heat treatment. The same figure shows also the results of three samples from powder-HIP block. Due to porosity found in the material, there is some deviation on the samples.

Table 2 Tensile requirements of CERN for raw material (not heat treated) copper and results after the HIP-treatment.

<table>
<thead>
<tr>
<th></th>
<th>CERN specification for raw material (no heat treatments)</th>
<th>HIP-treated samples (different pressures)</th>
<th>HIP-compacted copper from powder</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tensile strength Rm</td>
<td>240 - 280 N/mm²</td>
<td>HIP2: 211 N/mm²</td>
<td>HIP5: 220±4 N/mm²</td>
</tr>
<tr>
<td></td>
<td></td>
<td>HIP3: 206±7 N/mm²</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>HIP4: 206±2 N/mm²</td>
<td></td>
</tr>
<tr>
<td>Yield stress Rp 0.2%</td>
<td>200 - 240 N/mm²</td>
<td>HIP2: 121± N/mm²</td>
<td>HIP5: 51±3 N/mm²</td>
</tr>
<tr>
<td></td>
<td></td>
<td>HIP3: 109 N/mm²</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>HIP4: 56 %</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>HIP5: 40±13 %</td>
<td></td>
</tr>
<tr>
<td>Elongation at break A5 %</td>
<td>&lt; 25%</td>
<td>HIP2: 56 %</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>HIP3: 43, 44 %</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>HIP4: 44, 43 %</td>
<td></td>
</tr>
<tr>
<td>Hardness HV10</td>
<td>No requirement, 110 HV10 is normal for stock cold worked Cu</td>
<td>HIP2: 25.5±1.1 HV 10</td>
<td>HIP2: 30.2±0.5 HV 10</td>
</tr>
<tr>
<td></td>
<td></td>
<td>HIP3: 26.2±1.1 HV 10</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>HIP4: 25.3±1.2 HV 10</td>
<td></td>
</tr>
</tbody>
</table>

4.1.2. The effect on grain size

CERN specification for raw material is grain size ≤ 90 μm (in accordance with the standard ASTM E112-96 (2004) e1). Clearly, the high temperature of HIP-cycle will increase the grain size of a copper. From the samples below, one can see that the grains have been grown to mm/cm size. Still, some areas have rather small grains especially after HIP 3. The big grains are heavily twinned as can be seen in both macro and micro structures. Altogether, the microstructures are very typical for heat treated copper.
The grains are mostly mm size with some smaller grains between.

Figure 9 HIP2

The grains are mostly mm-size. However, there are areas with much smaller grains as shown right.

Figure 10 HIP 3

The grains are mostly mm-size with some smaller grains between.

Figure 11 HIP 4

4.1.3. The effect on surface topography

Surface topography of CLIC-components in the accelerator is important since electrical breakdowns are the main reason of the degradation of the components around the iris area. The breakdowns tend to concentrate on the areas with surface roughness due to the fact that pointy or rough surface areas can act as field emitters. Therefore, the components are diamond turned to very good surface quality. However, after machining, the components are assembled by diffusion bonding in high temperature and this treatment produces deterioration of the surface quality. The surface topography was investigated by Veeco scanner, optical microscope and SEM.
The difference between the three samples in Figure 12 is the pressure used as indicated in Table 1. In general, all three samples show the same features although the sample HIP3 with lowest pressure is somewhat different from the other two. In all samples, grain boundaries are grooved in from the surface plane and can be easily detected in metrological VEECO-scans and SEM-images. In addition to grain boundaries, also prior grain boundaries are well visible in the samples. The prior grain boundaries are most pronounced in HIP3-sample with lowest pressure.

![Image of three samples](image)

Figure 12 Discs after the HIP-cycles showing surface topography without etching. In HIP 3, most prior and present grain boundaries are visible giving an impression of very small grained sample.

Figure 13 shows rather clearly that pressure helps to keep the surface smoothness during the heat cycle in argon. The reason for surface degradation is grain boundary grooving and faceting. These phenomena can be seen in SEM images in Figure 14 to Figure 19 where grooves are the depressions around grain boundaries and facets are the step structures found in some grains. Most grain boundaries grooved in all three pressures so the larger degradation of smaller pressure is mainly originating from faceting. Faceting mechanisms are not very well understood but the consensus is that the surface is transforming to lower energy state at the treatment conditions (gases, temperature, impurities). On the other hand, the physical mechanism behind the faceting phenomenon is related to bulk diffusion, surface diffusion and atomic evaporation. Due to this uncertainty, it is also very difficult to conclude the influence of pressure on faceting. However, it is known that high pressure slightly reduces bulk diffusion rate but how it affects the other possibly contributing mechanisms is not known.

![Image of 3D topography scans](image)

Figure 13 Comparison of 3D topography scans show that the surface degradation decreases with increasing argon pressure.

**Surface topography of HIP2 (250 bar)**

Surface topography of HIP2 shows grain boundary depressions but the prior grain boundaries are only faintly visible.
Figure 14 Surface topography of HIP2. The blue depressions are prior or existing grain boundaries. There are also some elevations around grain boundaries but they can be seen much better from 3D-images in Figure 13.

Figure 15 Surface topography of HIP2. Prior and existing grain boundaries are mostly depressed/elevated. Faceting is rare.
Surface topography of HIP3 (100 bar)

Figure 16 Surface topography of HIP3. The surface is showing most of the grain boundaries and prior grain boundaries in both depressions and elevations.

Figure 17 Typical surface topography of HIP3.
Surface topography of HIP4 (1000 bar)

Figure 18 Surface topography of HIP4. The surface looks remarkably smooth with only occasional grain boundary depressions.

Figure 19 Surface topography of HIP5 by SEM. Some of the grain boundaries are grooved from the machined plane and also some prior grain boundaries are visible. There are grains that are heavily faceted.

4.2. Diffusion bonding tests

The diffusion bonding capsules described earlier were sawed in two halves and the cross sections were investigated with optical microscope. The images are compiled from about 15 images by a digital microscope. The thickness of the doughnut disc was 15 mm being the initial distance between the joints. The central cavity has been shrunk considerably due to the applied pressure.
HIP-parameters were chosen from the studied first HIP-cycle (HIP 1 with 1000 bar) that produced only mediocre deformation at full pressure. As can be seen from the results of HIP4 (Error! Reference source not found.), the results with the same parameters (HIP1 and HIP) are completely different and HIP4 produced completely closed central cavity. Therefore, the conclusion would be that there has had to been a leak in the capsule that has happened rather late in the process cycle.

The results considering the diffusion bonding tests show that with the used pressures there were always a good bonding achieved. This was investigated by microscopy showing some grains crossing the joint indicating a good bonding quality. Considering the surface quality, no difference was noticed between the joints. Since the inner cavity was deformed, the joint was also following the deformation of the discs. The deformed joints are marked to the macro images in Error! Reference source not found.Error! Reference source not found. whenever detected. One can notice that near the central cavity where the deformation is largest, the grains have been recrystallized during the treatment and the joint is very hard to be found. On the other hand, the joint areas away from central cavity did not deform and is more comparable to normal diffusion bonding.

Figure 20 HIP2 0.1 Ra. In close up, it can be seen that a grain is crossing the joint indicating good bonding.
Figure 21 HIP2 0.2 Ra. Again, good bonding achieved with this set-up.

Figure 22 HIP2 Ra0.8 Also the roughest surface utilized produces good bonding with HIP.

Figure 23 Left: HIP3, due to deformation and recrystallization the joint is hard to be distinguished especially near the central cavity. Right: HIP 4 at 1000 Bar, the opening in the middle has completely vanished except of the vertical drilling.

4.3. Powder compaction

Powder compaction by HIP is an established method for making near net shape components or bare raw materials. For CLIC-structures, only oxygen free copper is used to ensure that the hydrogen atmosphere used for diffusion bonding does not cause any adverse reaction with oxygen of the
copper. In case of atomized powder, production of very low oxygen gas atomized powder is very difficult. For this experiment, standard gas atomized powder with 720 ppm oxygen was used. This is far away from current CERN specification of 5 ppm but the price for purer powder would be very high if available. Thus, to study the properties of HIP-compacted copper a rather large capsule of \( \Phi 114 \times 200 \) mm was filled with copper powder, sealed by welding and degassed to vacuum normally (Figure 24).

A second experiment was conducted with the aim to experiment copper powder compaction utilizing a mould. Complex moulds are seldom used with HIP since the removal of the mould after HIPing is rather difficult. However, simple moulds to assist geometrical accuracy are sometimes utilized and, in case of copper powder, employment of a solid steel mould would be the most straightforward way. Similar to the diffusion bonding experiments, thick jacketed capsule design was used for this also. A negative of accelerator disc was machined from stainless steel AISI 316L as shown in Figure 25. There was also a thicker cover base plate inside the thick jacket in order to distribute the pressure axially. Both ends were then sealed by welding the thin cover plates (one with suction tube). After the previous steps, the capsule was degassed normally. The structure after opening can be seen in Figure 31. Compactions of the two test pieces were processed according to Table 1.

**Figure 24** Large powder compaction capsule and the result from the HIP-furnace.

**Figure 25** Drawing of the stamp disc (mould) and a sketch of the capsule in plots.
4.3.1. Large bulk cylinder

The cylinder (in Figure 26) was cut in two halves. One can notice the large shrinkage of the canister probably due to non-ideal powder size distribution.

Figure 26 Powder capsule after HIP-treatment and cut to two halves.

The canister was cut for further testing and a metallurgical sample were cut, polished and etched for examination. In order to measure oxygen level of HIP’ed sample, chips were machined dry and sent to Metalor for analysis. The result was 847 ppm which is only slightly increased from 772 ppm measured by the producer of the powder. This would indicate that the basic handling in air did not increase the oxygen content considerably. However, this result is not necessarily applicable to very low oxygen levels when more cautious powder handling may be needed.

Figure 27 shows a macro image of the HIP-compressed powder sample before etching and a larger magnification image of a pore found abundantly on the surface of the polished sample. Pores are very rarely found in HIPed materials and HIP-treatment is even used for removal of the porosity of castings. In order to stay open in high pressure HIP-treatment, there must have been an internal pressure in the pores in order to not being shrunk away. The origin of the porosity cannot be established entirely but most probably it is related to the high oxygen content and some reaction with other contaminants. The well-known reaction with oxygen and hydrogen producing water vapour porosity (hydrogen disease) in high oxygen content copper may be the responsible reaction. Since the starting point is powder, there is a very large surface area for any contaminations. Also, during the compaction there are natural cavities that can be filled by gas if generated during the processing. This is in contrast to solid material that needs to form those cavities. The effect is brittleness of solid copper due to oxygen/hydrogen reaction. On the other hand, the capsule was evacuated to high vacuum which is an effective way to remove any moisture that contains hydrogen.
The grain size of HIP samples compacted from powder were measured according to comparison method of ASTM E112 the grain size in G number is 6-7 (30-45 µm), which is smaller than the typical CERN stock copper with G number of 4-5 (60-90 µm). The hardness of powder HIP sample was 30 HV10 that is similar than dead soft annealed state of copper. Tensile test samples were prepared and the results are given in Table 2 together with solid copper tensile test results.

4.3.2. Near net shape preform
The capsule (the design is shown in Figure 25) was dismounted with considerable effort. First, the two 1 mm thick cover-discs were removed rather easily by turning and hammering. After the removal of the end covers, disassembling of the inner parts were attempted by pressing the thick jacket. Tensile test machine was utilized for this task but no movement out of the capsule were detected even with using 100 MPa maximum pressure. After that, two cuts were done (about 80% of the thickness of the thick capsule) to release the compressive forces and the capsule was again put to the press (this time normal industrial press). The parts inside were finally released but the rest of the jacket was now also broken as can be seen Figure 29 together with other disassembled parts after this stage. The other end of the mould (cover plate) was not attached to the other parts and detached apart from the copper by itself after the successful pressing.
Since the parts interested were still attached together, the rim of the disc was first cleaned with abrasive paper to see the junction. Since anti-sticking ceramic paint was used (it was noticed that it works well) the copper/stainless steel disc were hammered manually. After some effort, the copper and steel parts separated as shown in Figure 30. The whole assembly can be seen in Figure 31.

As can be seen from the figures, even though the shape of the component was achieved there is still a need for machining. Since rough machining of copper is very easy, the amount of material removed is not an issue and the biggest costs come from the fine diamond machining needed for this component. Therefore, this concept, although working as intended, is not very promising for future usage. For some other application, it may be a valid production method.

Figure 29 Left: the parts after removal of the cover, right: parts after cutting of thick capsule and press.

Figure 30 The junction of copper and steel before hammering and the two parts finally separated.
Figure 31 the assembly of the parts as they were in the capsule.

4.4. Brazing study

As stated in introduction, the aim of the brazing studies was to investigate the feasibility of utilizing a brazing alloy from electrodeposited layers of gold and copper in-situ by diffusion. If one studies the phase diagram of Au-Cu in Figure 32 it can be seen that the eutectic point is at 80.1 wt% of Au. Hence, this point would provide the easiest brazing operation and the lowest melting point achievable with Au-Cu. However, gold is a very expensive component of this alloy and a leaner alloy with gold would be appreciated. Red line in the phase diagram represents a composition that would be achieved if all deposited layers of gold and copper are co-diffused and mixed perfectly.

Figure 32 Au-Cu phase diagram [5]. The red line is the composition of fully and evenly diffused Au- and Cu-layers (63 wt.% of Au) only counting the deposited layers.

This study was a continuation to earlier experiments [6], [7]. In those experiments, only gold was electrodeposited on one joint wall. In the first experiment, the temperature was set at 900°C that is below the eutectic temperature of Au-Cu system. Therefore, it was not possible that there were any liquid formed during the processing and the brazing failed. In the second experiment, the
temperature was increased to 930°C that is above the eutectic point. In principle, it is then possible that some liquid is forming. Still, the diffusion starts from the gold-copper interphase that is inside the solid Au-Cu system. Therefore, it is possible that at one time, only rather thin layer of the copper enriched coating is at liquid state. Then, when the diffusion reaches the edge of the coated part, only very thin layer would be available to produce the brazing. This is also helped with the fact that diffusion starts already at the heating up of the sample although the rate of diffusion is much slower at lower temperatures. It is then possible that at the time the piece arrives at 930°C, the coating is already diffused to the stage that it is not possible to produce eutectic composition.

Since electrodeposition makes a very good bonding to the base material allowing diffusion through the boundary, a nickel layer was introduced to eliminate or at least hinder the gold diffusion to the base copper. In order to examine the diffusion behaviour in the brazing, three different samples were coated with the layer systems given in Table 1.

Table 3 Details in brazing experiments. Three sets of layer systems were manufactured at CERN by electrodeposition (all thicknesses in µm):

<table>
<thead>
<tr>
<th>Layer</th>
<th>Set 1</th>
<th>Set 2</th>
<th>Set 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni diffusion barrier</td>
<td>10</td>
<td>10</td>
<td>10</td>
</tr>
<tr>
<td>0th Cu layer</td>
<td>25</td>
<td>20</td>
<td>17.5</td>
</tr>
<tr>
<td>1st regular Au layer</td>
<td>20</td>
<td>10</td>
<td>5</td>
</tr>
<tr>
<td>1st regular Cu layer</td>
<td>5</td>
<td>2.5</td>
<td></td>
</tr>
<tr>
<td>2nd regular Au layer</td>
<td>10</td>
<td>5</td>
<td></td>
</tr>
<tr>
<td>2nd regular Cu layer</td>
<td></td>
<td></td>
<td>2.5</td>
</tr>
<tr>
<td>3rd regular Au layer</td>
<td></td>
<td>5</td>
<td>2.5</td>
</tr>
<tr>
<td>3rd regular Cu layer</td>
<td></td>
<td></td>
<td>5</td>
</tr>
<tr>
<td>4th regular Au layer</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Total thickness</td>
<td>55</td>
<td>55</td>
<td>55</td>
</tr>
</tbody>
</table>

The layered copper discs and four neutral cover discs were cleaned, pickled and stacked according to the CERN instructions. The stack was then treated in a high vacuum furnace Torvac at VTT in Finland. The set-up is given in Figure 33. The dimensions of the discs were Ø40 x 15mm. After degreasing the discs were rinsed with water and ethanol and dried with hot air blow (hair dryer).

Brazing program with vacuum level of $2 \times 10^{-4} - 7 \times 10^{-5}$ mbar was utilized as follows:

- Ramp 20 - 950°C at the rate of 20°C/min
- Hold 1 hour
- Ramp to 1000°C at 100°C/h
- Hold 10 minutes
- Cooling free with furnace

After brazing the stack was examined visually. The cover discs had the shiny appearance of copper and the coated discs had matte appearance resembling sand blasted surface. The surrounding of the brazing joints had the roughest appearance. For some reason, the alumina plates under the stack and weight were broken as can be seen in Figure 34 (showing also the numbering of the joints). The brazed stack was cut by spark erosion in two halves and metallographically polished.

The joints were investigated by SEM that led to several findings. Firstly, it was found that there was replacement of material that filled some of the cracks/discontinuities. The material probably stuck only in the small crack openings. Some images are presented in Figure 35 and Figure 36 showing the unsound structure at some parts of the joints. One can also see the presence of gold at the vicinity of the joints as lighter areas.

Figure 34 Left: naming of the joints and right: the copper disc stack after brazing. The alumina plates under the stack and steel weight were broken during the treatment.

Figure 35 Images showing the joint areas with transferred grinding or cutting material.
Figure 36 Two images of the same area with two magnifications revealing the unsound structure of the joints on some locations. The image on the right shows how there has been a collapse of the thin layer of displaced grinding material. One can notice the grinding scratches on the piece buried in the crack.

Bearing in mind the results of the SEM-observations, the quality of the brazed joints was investigated by an optical microscope. The results are presented in Figure 37 showing the discontinuities of the joints in red. One should notice that the two best joints (J2 and J6) both have excess brazing material at the chamfered outer rims of the joint. This could indicate that there has been lack of liquid brazing material on other joints. In Figure 37, there are also markings of the EDS analyses made across the joints marked as SOI# (site of interest). As the chamfers are filled with gold containing material, it can be concluded that there has been liquid material present during the brazing cycle. This can be confirmed also by investigating Figure 38 that shows a shrinkage cavity at one of the filled chamfers.
Figure 37 Macro images of the joints marked as in Figure 34. Red markings show the places with gaps. Site of interests are marked in the macros and correspond to SEM/EDS scans below.

Figure 38 SEM images of the chamfered outer rims of the joints 2 (filled with brazing material) and for comparison, joint 4 (no filling, although a good joint near the chamfer).
**Diffusion at the joints**

The diffusion behaviour of the coatings and surrounding material is of great importance since this is the base of the intended brazing action. Diffusion was investigated by quantitative EDS-analysis that is very good tool for this investigation. Also, as it was seen on the images earlier, elemental contrast in SEM-images can reveal diffusion in qualitative fashion. The contrast between gold and copper is such that either secondary electron detector or back scattering detector can be used but distinction between nickel and copper is not possible. In Figure 40, there are two images showing elemental distribution of good and bad joints. One can also see the difference between the sides of the joints; the sharper elemental change (change in darkness) is the side of original coating with nickel.

![Image of secondary electron images showing diffusion](image1)

In few spots there were also situations where the gold had clearly diffused to the other side of the opening as shown in Figure 40. The explanation for this was not revealed in our investigations since the edges of the cracks are not showing any sign of smooth surfaces indicating molten stage edge separation. However, it is possible that the crack edges are somehow damaged during sampling or polishing.

![Image of an opening showing peculiar gold diffusion](image2)

EDS analyses of Nickel (Ni), copper (Cu) and gold (Au) were conducted and results are shown in Error! Reference source not found.Error! Reference source not found.. One can again notice various pes of joint qualities achieved. The locations of the scans were selected so that they represent all joints and also visually distinctive joint types. Basically, two kinds of joints were examined: those...
with smooth transition of brightness across the joint and those with sharp transition. At the time of the EDS-scans it was not realised that the locations with a sharp boundary of gold diffusion are not necessarily good joints but can be just a result of polishing of the sample. The images and the graphs are synchronized so that the white dot-line is showing the actual measurement line location and the X-axis of the graph is then normalized to the same length. If one studies the scans in more detail, there are several interesting findings:

1. The nickel layer can be easily found by EDS
2. If the nickel line shows relatively high peak in the graph – then the gold diffusion has been effectively blocked as shown in Figure 41a.
3. In case of low nickel peak, the gold distribution line shows only little error on symmetricality in X-axis.
4. If the nearby graphs with good and bad joints are put in the graph, one can see clearly that the sides that the materials were deposited are nearly identical. Hence, the good joint has extra material to produce a good joint.
5. As shown in Figure 43, all measurements of good joints show very similar compositional distribution in spite of different layer systems.

Figure 41 a) EDS-scan at well diffused area and good joint (joint 1, SOI 1), b) this side of the disc have a thicker Ni-layer effectively blocking diffusion of gold (joint 2, SOI 3).

Figure 42 a) Joint 3 with good (smooth transition of elements) and bad (sharp transition) joint areas, b) the EDS-scans of SOI 5 and 6 are put in the same graph and scaled showing that bad joint areas have substantially less total gold.
Discussion on the brazing study

In the present study, a nickel layer was introduced to prevent excessive diffusion. In theory, it was assumed that nickel would prevent diffusion and the layers of Au and Cu would interdiffuse and produce a brazing alloy in lower temperature. Later, the piece would be brought to higher temperature for actual brazing. However, the diffusion treatment temperature was 950°C which is higher than the eutectic point. Hence the Au-Cu system will be partly melted during the diffusion treatment. It is possible that some of the melted alloy is relocated during the diffusion treatment. This relocating liquid must be high in gold since otherwise it would not be melted. The result would be that the joint areas that gained the liquid would have better chances to be brazed since there are more material available and the gold content is higher that provides lower melting temperature. The areas that lost the liquid would be poor in gold and may have only thin liquid layer not capable of bridging the gap. This is of course hypothetical discussion but the facts can be studied by separating the diffusion cycle and measuring the actual starting coatings. Also mathematical methods are possible since diffusion phenomena can be rather accurately simulated. The calculations need to be solved numerically though.

Electrolytic deposition is a method sometimes used for brazing and it can be used for instance to braze steel pieces by pure copper. In case of an alloy (instead of pure metal) needed for brazing it becomes more complicated. Still, if one makes the diffusion treatment below eutectic point of the alloy system, prevents the diffusion to the coated layers only and optimizes the brazing material, gaps, surface quality and so on, there is no reason why it should not be feasible joining method.

5. Conclusions

Hot Isostatic Pressing (HIP) is an excellent way to produce powder based components or it can be utilized for diffusion bonding of same or mixed materials. In case of copper powder, the high oxygen level is a big hindrance with the mostly utilized gas atomizing powder production. It could be seen in our sample with porosity although there is no conclusive evidence that it was caused by oxygen in the copper. Therefore, copper produced from powder by HIP-route do not give any benefits over conventional ones unless a cheap, low oxygen powder can be found. Then, in theory, it is possible to produce small grained raw material. It would still be more expensive than conventionally produced OFC-copper. The experiment considering HIPing with negative mould was successful. However,
considering the problems associated with powder and the difficult disassembly of the mould set-up, one cannot see much feasibility on utilizing it for accelerator structures.

HIP-technology is frequently used for diffusion bonding also. In theory, that could be a very good method also for copper-copper bonding. The main problem considering HIP-bonding and CLIC accelerator structure is that there is a cavity inside the structure. Since HIP-bonding is done with capsuling and evacuating the inside of the capsule, the cavities tend to shrink considerably or vanish altogether as was seen in experiments done. In theory, one could produce an arrangement with inside mould/removable substance that would support the inner cavity from the HIP-pressure, but feasibility over the current joining method of regular diffusion bonding would be non-existent.

Experiments with brazing were partly successful showing that it could be possible to use such a system with electrodeposited coatings. The experiment showed the unnecessarity to use fine layers with used treatment parameters and showed that with tweaking of the parameters, it should be possible to produce good bonding.

6. Outlook
Although some of the experiments with HIP-technology were successful, there were no indication found that any of them would be feasible for production of accelerator components made of copper.

The brazing experiments are continued at CERN with the aim to understand the roles of diffusion part and the actual brazing. Therefore, an experiment of diffusion treatment is planned and will be completed by heat treating the electrodeposited Ni-Cu-Au layers at 900°C.

7. Acknowledgement
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Bibliography