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2017 IOP Conf. Ser.: Mater. Sci. Eng. 181 012005

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Development of a Cu-Sn based brazing system with a low brazing and a high remelting temperature

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Abstract. Objective of the project presented is the development of a joining process for hot working steel components at low brazing temperatures leading to a bond with a much higher remelting temperature. This basically is achieved by the use of a Cu-Sn melt spinning foil combined with a pure Cu foil. During brazing, the Sn content of the foil is decreased by diffusion of Sn into the additional Cu resulting in a homogenous joint with a increased remelting temperature of the filler metal. Within this project specimens were brazed and diffusion annealed in a vacuum furnace at 850 °C varying the processing times (0 – 10 h). The samples prepared were studied metallographically and diffusion profiles of Sn were recorded using EDX line scans. The results are discussed in view of further investigations and envisaged applications.

1. Introduction

Braze filler compound systems that contain of at least two metallic components, for example in form of multilayer coatings or foils are characteristic for transient liquid phase bonding (TLP-bonding). The thermal joining process takes place by interdiffusion of the components resulting in a homogeneous filler alloy with a higher remelting temperature compared to the melting temperature of the pure filler metal. This, of course, depends on dwell times and temperatures applied to the TLP-system chosen [1].

In the present study this fundamental mechanism is used to create durable joints at moderate joining temperatures. As shown in figure 1, two Cu75-Sn25 foils (wt.%) were applied with an additional copper foil to join hot working steel components.

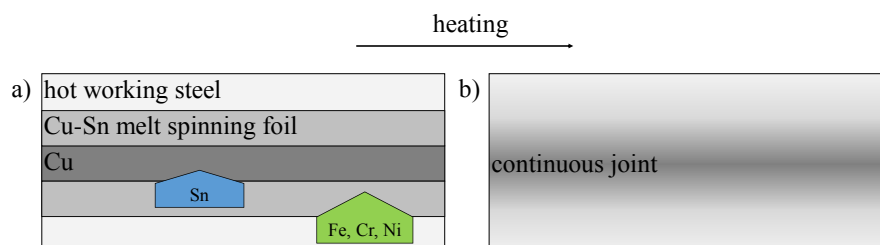


Figure 1. a) initial state of the brazing process; b) completed brazing process.

At first a conventional brazing process is performed, where the liquid Cu-Sn foil wets the steel and the copper surfaces. With increasing dwell time more and more copper is dissolved in the liquid braze metal, since an isothermal solidification of the braze metal takes place. Then a further diffusion heat treatment is performed in order to decrease the Sn content further by solid state diffusion. When

reaching a homogenized joining zone with the remelting temperature envisaged, the joining process can be stopped.

Objective of the described work is the investigation and description of the diffusion process as a function of the brazing parameters in order to determine the optimal process conditions for this kind of joining process. Prior to the experimental details a short introduction to the Cu-Sn brazing system and the mathematical description of Cu-Sn interdiffusion is given below.

Cu-Sn system

The binary alloy containing 75 wt.% Cu and 25 wt. % Sn (85 at.% Cu, 15 at.% Sn) was chosen for this study in the form of a melt spinning foil with a thickness of 40 μm. It was supplied by the company Vacuumschmelze. As shown in figure 2, the brazing process was performed at 850 °C varying the processing times (0 – 10 h).

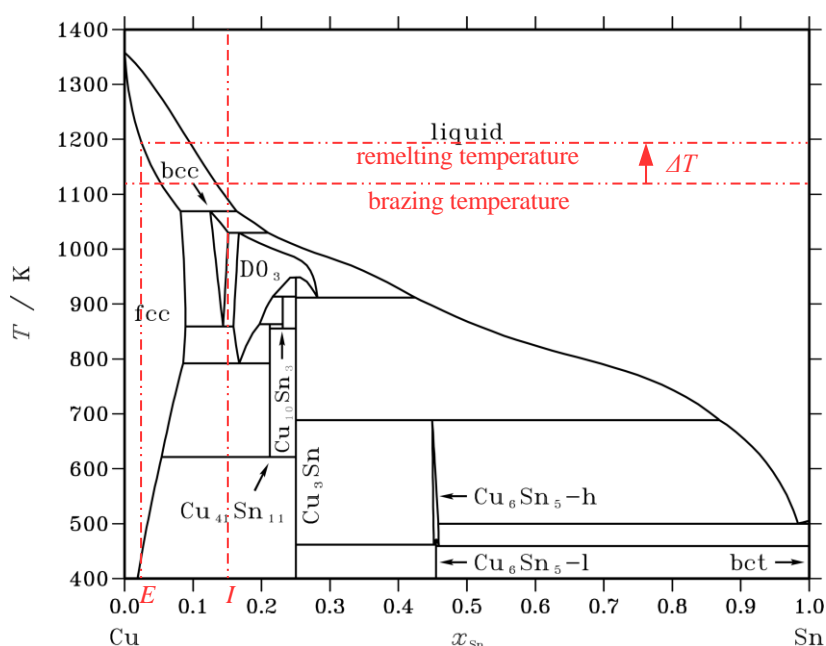


Figure 2. Cu-Sn phase diagram [2].

During the brazing process the initial composition I changes to a composition E with a decreased Sn content by the diffusion of Sn into the additional copper foil, which causes a shift ΔT in melting temperature.

Diffusion processes

For the present brazing system, the composition within the resulting joint is determined by the brazing time and temperature. Therefore a mathematical description of the diffusion processes is essential to understand the nature of the joining process and the resulting microstructure of the brazed seam and to forecast appropriate process conditions for envisaged applications.

The temporal evolution of a given concentration distribution c is described by the diffusion equation

$$\frac{\partial c(x,t)}{\partial t} = \frac{\partial}{\partial x} \cdot D(c, t) \cdot \frac{\partial c}{\partial x}(x,t) \quad (1)$$

with the diffusion coefficient $D(c,t)$.

Analytical solutions do exist in special cases, particularly for a constant diffusion coefficient [3]. But in the present case there is no analytical solution available since the interdiffusion of copper and tin depends highly on the actual tin concentration. Hoshino et al. [4] studied the diffusion of Sn in the Cu-Sn system intensively. Based on their data the concentration and temperature dependence of the diffusion coefficient of Sn in Cu can be expressed as

$$D_{Sn\ in\ Cu} = D_0 \cdot \exp(-E/RT) \cdot \exp(a \cdot c) \quad (2)$$

with

diffusion coefficient $D_0 = 1.45 \cdot 10^{-5} \text{ m}^2/\text{s}$,
 activation energy $E = 179.8 \text{ kJ/mol}$,
 universal gas constant $R = 8.314 \text{ J/mol K}$,
 temperature $T = 973 - 1125 \text{ K}$,
 and concentration coefficient $a = 0.269$.

This data were used to solve the diffusion equation numerically for the boundary conditions given by the geometry and brazing conditions of the samples processed in this work.

2. Experimental details

Filler metal

The filler metal 75Cu-25Sn was supplied by the company Vacuumschmelze as a melt spinning foil with a thickness of 40 μm . It was used with a pure copper foil with a thickness of 500 μm as a filler system (figure 1).

Base metal

Hot working steel 1.2714 (A681 L6) was used as base metal for the joining process. Its composition is shown in table 1. This material is an air-hardening steel and it is typically solution annealed with the same temperature, which is used here for the brazing and diffusion treatment of the joint. So the joining process may be coupled with the hardening of the material in one furnace process.

Table 1: Composition of the base metal used.

1.2714	C	Cr	Mo	Ni	V
content in wt.%	0,55	1,10	0,50	1,70	0,10

Heat treatment

The samples studied were brazed in a vacuum furnace at 850 °C varying the processing times (0 – 10 h). Figure 3 shows the temperature during the heat treatment of the specimens as measured using companion specimens instrumented with thermocouples.

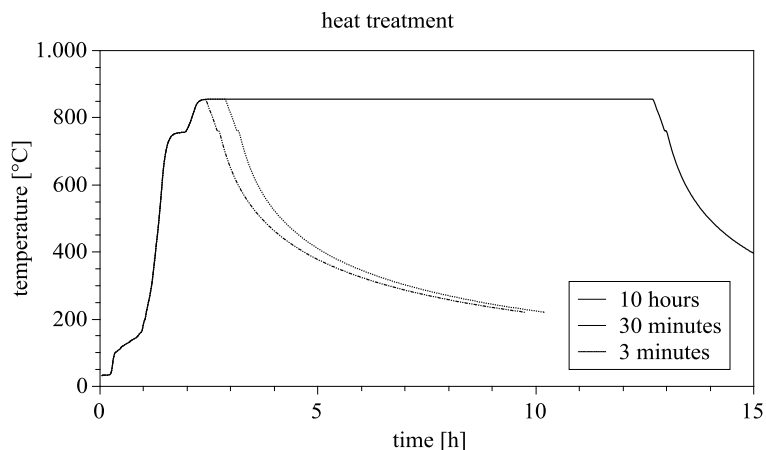


Figure 3. Temperature during heat treatment.

Interaction between melt spinning foil and additional copper

Joining using the filler system employed requires a fundamental understanding of the interaction between the Cu-Sn foil used and the additional copper upon heating, which can be studied best on samples with simple and defined joining geometries. Hence the diffusion of Sn in Cu was studied on brazing gap specimens shown in figure 4.

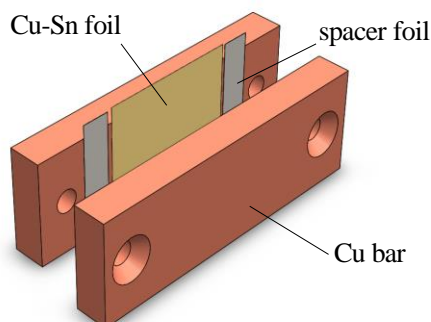


Figure 4. Cu gap specimen.

Two copper bars (45 mm x 15 mm x 5 mm) were held together by stainless steel screws. Stainless steel foils with a thickness of 40 μm were used as spacers and placed in between the copper as well as the Cu-Sn melt spinning foil. Once the brazing process was performed, the surface was polished to investigate the distribution of elements within the brazed joint by means of EDX line scans.

One set of specimens was brazed for 3 minutes at 850 $^{\circ}\text{C}$ to investigate the interaction between melt spinning foil and additional copper during the melting and isothermal solidification process. No diffusion annealing was performed.

In a further step the influence of annealing on the resulting Sn concentration was investigated. Therefore brazing processes were performed for 30 minutes and 10 hours.

Brazing hot working steel specimens

The behavior of the brazing system in the case of application is of special interest. Hence specimens corresponding to figure 5 were brazed and investigate similar to specimens mentioned in section 2.4.

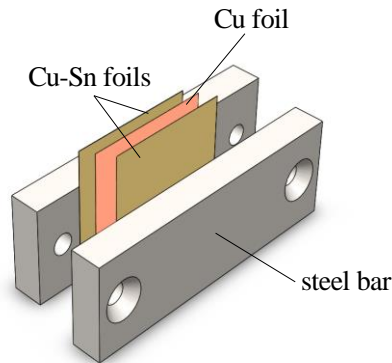


Figure 5. Hot working steel specimen.

Two hot working steel bars (45 mm x 15 mm x 5 mm) were held together by stainless steel screws. Two pieces of melt spinning foil (40 μm) combined with a copper foil (500 μm) were placed between the hot working steel as a stack prior to brazing. Metallographic cross sections were prepared from the brazed specimens in order to investigate the microstructure of the brazed seam as well as the distribution of elements within the brazed joint by means of EDX line scans.

3. Results

Interaction between melt spinning foil and additional copper

The left picture in figure 6 shows the microstructure of a Cu/Cu-Sn brazing joint specimen after brazing for 3 minutes at 850 $^{\circ}\text{C}$. On cooling to room temperature Cu_3Sn phases formed in the centre of the brazed seam. These phases did not appear, if the joining process was performed for 30 minutes (middle image in fig. 6) due to the advanced dilution of Sn within the brazed seam.

After brazing at 850 $^{\circ}\text{C}$ for 10 hours the brazed seam was homogenized and some small pores are visible now in the centre (right image), which is the well known Kirkendall effect, resulting from solid-state interdiffusion of atoms of different size.

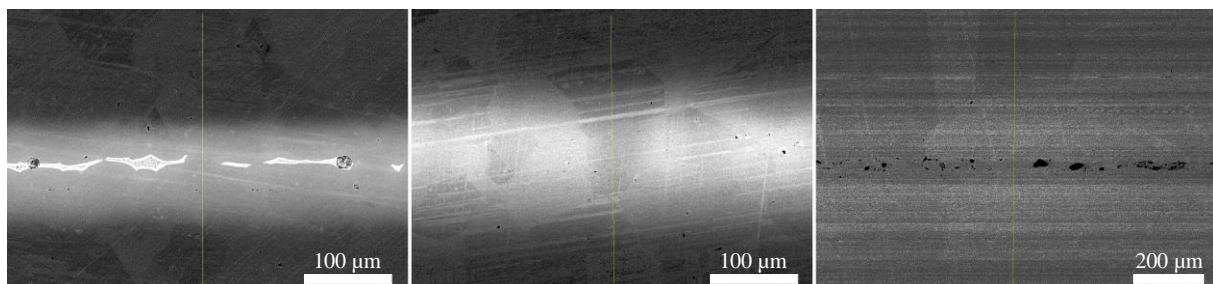


Figure 6. Microstructure of Cu specimens brazed at 850 $^{\circ}\text{C}$ using Cu-Sn melt spinning foil. After 3 minutes (left), 30 minutes (centre) and 10 hours (right).

A closer look at the evolution of the Sn diffusion profile throughout the joining zone is given by EDX line scans shown in figure 7. The rectangular plot illustrates the start condition corresponding to the melt spinning foil with a Sn content of 15 at.%. In the following measured diffusion profiles of Sn in Cu are compared to calculated profiles.

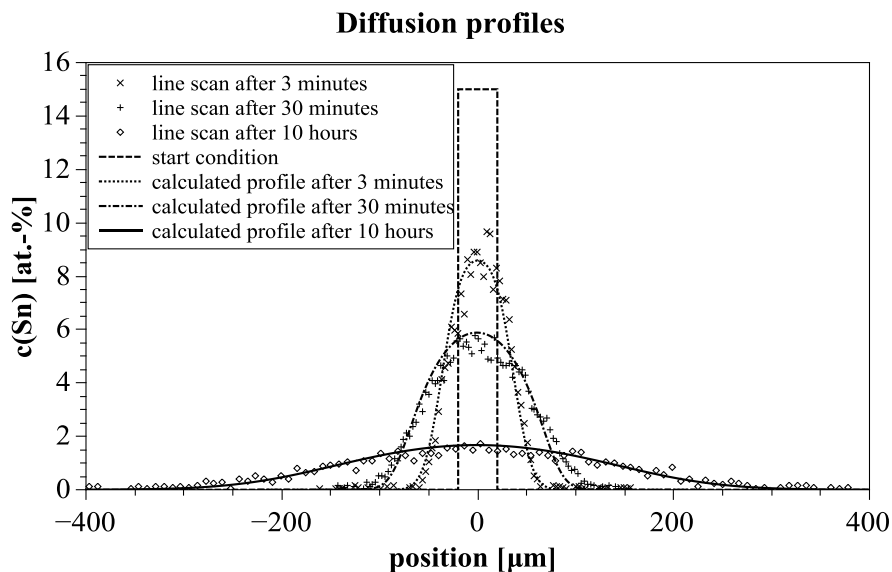


Figure 7. Diffusion profiles of Sn in Cu after several processing times.

In all cases there is a good match between the measured and calculated profiles, so the observed diffusion of Sn in Cu agrees well with the numerically solved model when using the diffusion coefficient given in equation (2).

Brazing hot working steel specimens

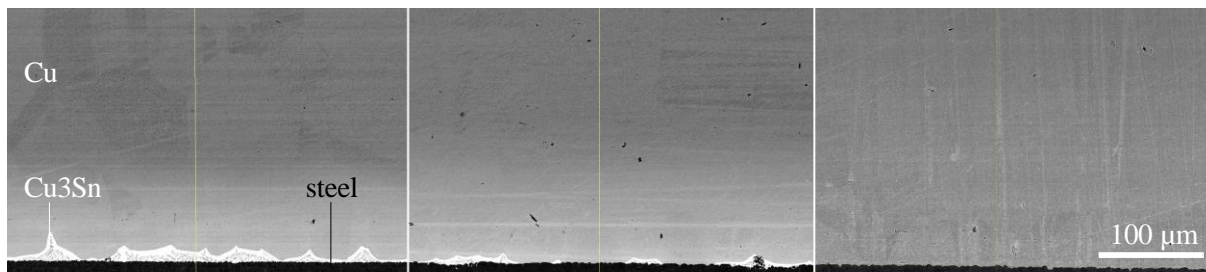


Figure 8. Microstructure of hot working steel specimens brazed at 850 °C using Cu-Sn melt spinning and copper foil. After 3 minutes (left), 30 minutes (centre) and 10 hours (right).

The left picture in figure 8 shows the microstructure of a hot working steel brazing joint specimen after brazing for 3 minutes at 850 °C. On cooling to room temperature Cu_3Sn precipitations formed at the interface to the steel. These precipitations did not appear that distinct, if the joining process was performed for 30 minutes (middle image in fig. 8) due to the forced dilution of Sn within the brazed seam. After brazing at 850 °C for 10 hours the brazed seam was homogenized (right image).

A closer look at the evolution of the Sn diffusion profile throughout the joining zone is given by EDX line scans shown in figure 9. The positions of the Cu-Sn and Cu foil prior to brazing are indicated by vertical lines. Depending on the annealing time the maximum Sn concentration at the steel interface decreased and therefore the remelting temperature increased. After 30 minutes of annealing the remelting temperature already is 930 °C according to the solidus temperature of a Cu alloy with max. 5 at.-% Sn content, see figure 2. An annealing for 10 hours yield to a further decrease of the Sn content to finally ca. 2 at.-% which corresponds to a solidus temperature of 980 °C.

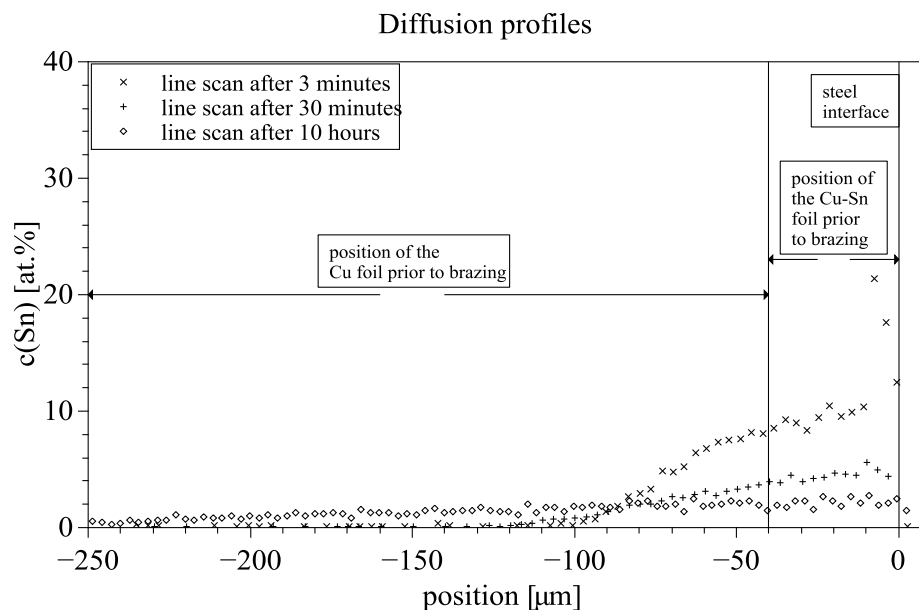


Figure 9. Diffusion profiles of Sn in hot working steel specimens after several processing times.

4. Conclusion and outlook

The Cu75-Sn25 melt spinning foil in combination with pure copper as additional solid filler metal is a promising candidate for joining hot working steel via TLP-bonding. Especially the high diffusion coefficient of Sn in Cu enables quite moderate brazing temperatures (850 °C) and annealing times (< 10 h) and results in homogenous, high melting copper based joints in a vacuum brazing process. Even after 30 minutes of brazing at 850 °C a remelting temperature of 930 °C was achieved and it can get even higher on further annealing.

In the ongoing studies the brazing time will be optimized with the aim of avoiding the formation of Cu₃Sn during the cooling process. So annealing times will be varied between 30 minutes and 10 hours until the minimum annealing time is found. Findings gained will be compared to numerical calculations.

In the final steps hot working steel specimens will be brazed and hardened in one furnace process and their technological properties like hardness, tensile strength and corrosion behaviour of the joint as well as the base material will be assessed.

Acknowledgment

The IGF Project IGF 18.706 N/DVS Number.: 07.081 of the research association "Forschungsvereinigung Schweißen und verwandte Verfahren des DVS, Aachener Straße 172, 40223 Düsseldorf" was, on the basis of a resolution of the German Bundestag, promoted by the German Ministry of Economic Affairs and Energy via the Federation of Industrial Research Associations (AiF) within the framework of the programme for the promotion of joint industrial research and development (IGF).

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