LOW TEMPERATURE JOINING FOR HEAT EXCHANGERS AND OTHER APPLICATIONS

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The paper in hand presents a joining method, using tin and zinc based solders and a specific way of mechanical oxide removal. The fluxless process works in air and is carried out at temperatures of 250°C (482°F) and 450°C (842°F) respectively. Process and alloy features and example applications are given and joint structures and selected properties of similar and dissimilar joints between aluminum, copper and steel are reviewed.

Keywords: fluxless soldering, low temperature joining, aluminum, copper, steel, ceramics, heat exchanger, porous materials, pvd targets

INTRODUCTION

Among constructive materials, aluminum, copper and their alloys on one hand and steel on the other hand have gained the biggest technological importance. In producing components, the effectiveness of the individual advantages of these materials classes can be very often enhanced by combining these materials with each other within hybrid structures. In most cases, the optimum use of the materials can be achieved only, if a joining technique is applied that fits the different materials and meets the demands of the service conditions (Ref. 1). Among material joining techniques, soldering and brazing are widely used, due to their high degree of flexibility. Besides the suitability of the process for a certain part geometry, it is necessary to adapt the joining method to the material or material combination to be joined respectively (Ref 2). Conventionally soldering and brazing operations use fluxes for removing surfaces oxides. These processes are carried out otherwise in protective atmosphere or under vacuum. Because of these disadvantages the objective has been set to develop and establish a soldering process that can be performed in air without using any fluxes. In order to increase the process reliability when aluminum materials with solidus temperatures close to 600°C (1112°F) are joined, a soldering temperature below 500°C (932°F) is desired. The removal of filler metal and base material oxides occur mechanically.

PROCESS

Process Fundamentals

The joining process is comprised of two major stages which are heating up of the parts to be joined and oxide removal, **fig. 1 (top)**. During heating up of the parts, oxides are formed on both base material and solder alloy as well. These oxide scales impede wetting. On reaching the soldering temperature, the surface oxides get broken mechanically and the molten solder has direct contact with the base materials. Due to reactive elements in the solder (see below) a wide range of materials, including ceramics and glasses, can be wetted.

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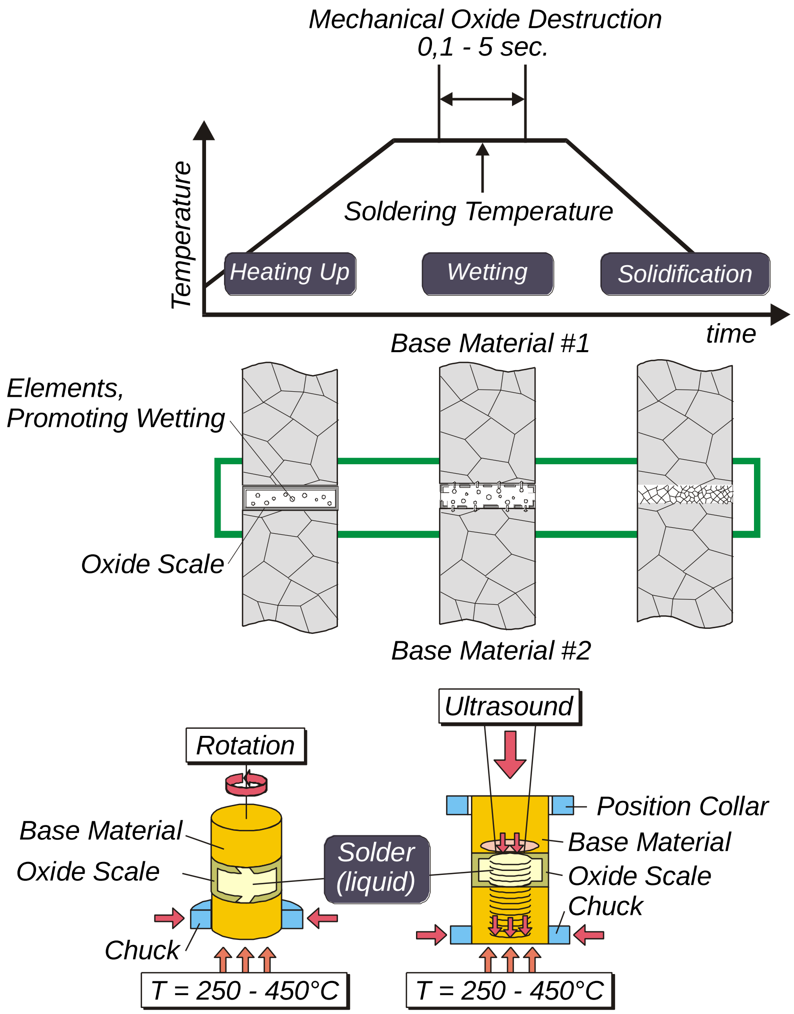

Fig. 1: Soldering principle with mechanical oxide removal and methods of oxide destruction

The heating is done by flame, hot air or induction heating respectively. The method used for destroying the oxide scale on the liquid solder depends on the geometry of the parts to be joined. Two principles have been generally employed, **fig. 1 (bottom)**. For smaller parts ultrasound is induced into the top part. Vibrations propagate through the base material and destroy the oxides at the interface between liquid solder and base material by cavitation. The oxides get finely dispersed in the solder and are partly pressed out of the gap to be filled. The small oxides act as grain center during solidification and cause refinement of solder zone with enhanced mechanical properties. For bigger parts, friction is favored instead of ultrasound. In the case of joining cylindrical parts the bottom part is fixed and the top part is pressed towards the bottom part and rotated. The friction between liquid filler metal/filler oxide and the base material destroys the oxide scale and wetting takes place.

FILLER METALS

The novel solder alloys and the soldering process has been developed to industrial standard by Euromat GmbH in collaboration with the Materials Science Institute in Aachen (Ref 3-5).

The basic structure of these solders consists of five elements. The major constituent, which mainly influences the melting range, is tin or zinc respectively. Most alloys contain titanium as second element. The elements silver and/or copper results in a reduction of surface tensions in molten state and contribute to strength enhancement of soldered joint. Further additions are cerium or a mixture of rare earth elements and gallium. Owing to its high affinity to oxygen cerium reacts with oxygen from the surrounding air. During solidification the oxide particles which act in the first place as grain centers and form barriers for grain growth result in a refined structure with increased strength behavior. Gallium promotes wetting on metallic materials. The gallium content need to be kept low to avoid the formation of low melting eutectics.

EXPERIMENTAL PROCEDURE

Solder Alloys

For investigation purposes six solders have been selected, **tab. 1**. The melting range of these alloys have been determined by thermal analysis (DTA).

Alloy #	Composition		Melting Range [°C/°F]
	Constituents [wt.-%]	Additions[wt.-%]	
1	Sn Ag4 Ti4	Ce, Ga < 0,5	220 - 229 / 428 - 444
2	Sn Ag3 Cu1 Ti4		215 - 220 / 387 - 444
3	Sn In5 Ti4		208 - 238 / 406 - 460
4	Zn Al6 Ag6		380 - 395 / 716 - 743
5	Zn Ag4 Ti4 Cr0,7		415 - 432 / 779 - 810
6	Zn Ag4 Ti4 Ni0,3		415 426 / 779 - 799

Tab. 1: Solder composition and melting range

The solder alloys have been constituted under the boundary condition that the solder perform close melting ranges. The melting ranges are 208 - 238°C (406 - 460°F) for the tin based alloys and 380 -426°C (716 - 810°F) for zinc based alloys. The soldering temperatures are 250°C (482°F) or 450°C (482°F) respectively. For material characterization one tin based alloy (#1) and one zinc based alloy (#5) have been selected and physical data, which are relevant in terms of joining and application, have been ascertained.

Physical Property	Solder (#)	
	SnAg4Ti4CeCeGa (#1)	ZnAg4Ti4CeCeGa (#5)
Density [g/cm^3]	7,4	7,1
Electrical Resistance [10^{-7} Ω·m]	1,6 - 1,7	1,4 - 1,7
CTE [10^{-6} 1/K]	17,7	26
Thermal Conductivity [W/m·K]	48	81
Specific Heat Capacity [J/kg·K]	243	416

Tab. 2: Physical properties of solder alloys SnAg4Ti4CeGa (sold. #1) and ZnAg4Ti4Cr0,7CeGa (sold. # 5)

Base Materials

As base metals the non-hardenable alloy AlMg3, the hardenable alloy Al Cu2 Mg1,5 Ni (AA2618) and E-Cu57 have been used. Two types of steel have been additionally considered, i.e. structural steel S235 JR (AISI 1010) and stainless steel X5 Cr Ni 18 10 (AISI 304).

Soldering Process: Experimental Set Up And Procedure

The method of the presented fluxless soldering process is based on the mechanical destruction of oxide scales building up on liquid solder and parent material. The oxide of the base material gets partially cracked, so that a metallurgical interaction between solder and base metal constituents can take place. The solder melt undermines present surface oxides and leads to complete wetting of the base material. For this test series the application of the solder is a two step process. The first step is precoating of the samples to be joined with a specific soldering iron. The special feature of the soldering iron is a integrated ultrasound transducer system, that enables fluxless soldering by breaking solder and base metal oxides by means of vibrations, **fig. 2 (left)**.

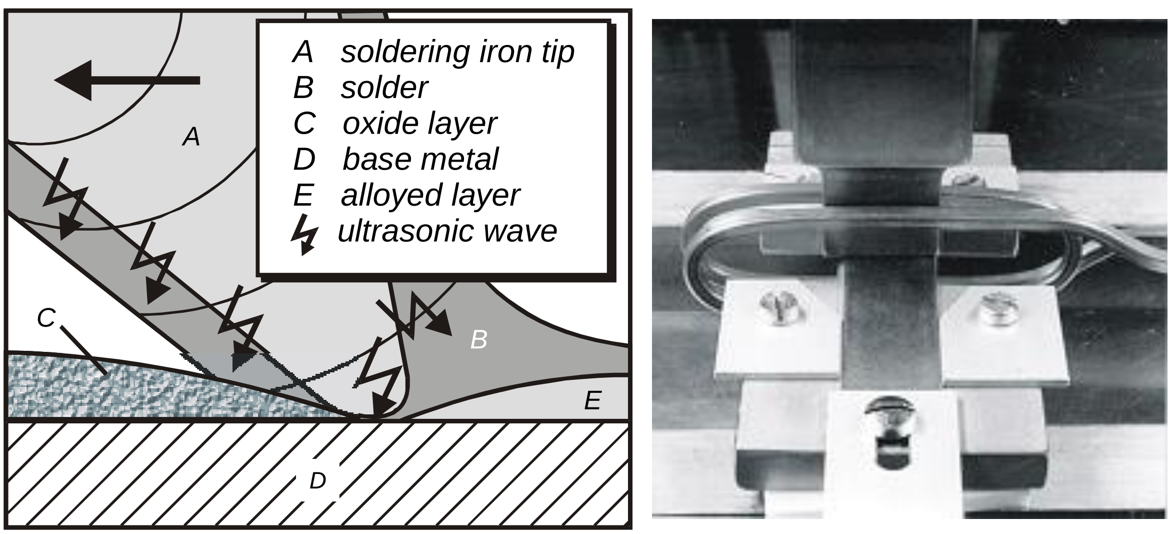

Fig. 2: Ultrasonic soldering iron (left) and induction coil with horn (right)

The oxide scales of base and filler metal are destroyed by ultrasound waves which are applied with a frequency of 60 kHz and an amplitude of 2µm. The temperature of the precoating process, which is equal to the joining temperature, is 250°C (482°F) for solder # 1 – 3, 420°C (788°F) for solder # 4 and 450°C (842°F) for solder # 5 and 6 respectively.

Samples for strength testing have been heated by inductive heating. With inductive heating eddy currents which result in resistance heating, are coupled without direct contact into the workpiece (Ref.6). For all tests a high-frequency transformer with a nominal wattage of 12 kW and a frequency range of 100 – 300 kHz has been used. On reaching the soldering temperature, workpieces are dwelled for 15 seconds at this temperature to achieve a homogeneous temperature distribution in the joining area. To produce shear strength samples, ultrasound has been applied perpendicular to the joining area, **fig. 2 (right)**. Ultrasound waves propagate through the sample and break the oxide layer. The vibration frequency was

20 kHz and the amplitude was 10 µm. The specimens were overlap joints with a joining area of 20 x 3 mm².

Tensile strength samples were produced by using a special machine, **fig. 3**. After heating up both parts to the soldering temperature with a heating plate, the top part was rotated with velocity of rotation of 100 R/min. The top part was simultaneously pressed on the bottom part with a force of 100 N for five seconds. During cooling down of the workpiece, the applied forced has been increased to 500 N. As samples rods with 15 mm diameter and 100 overall length were used.

Fig. 3: Special installation for fluxless soldering operation

TEST RESULTS AND DISCUSSION

The joint quality has been evaluated by light microscopy. For both low melting alloys (# 1 – 3) and higher melting fillers (# 4 – 6) a good wetting of the base material could be achieved. In case of joining E-Cu57 with solder # 1 (SnAg4Ti4CeGa) the specific intermetallic zone with the composition of Cu_3Sn/Cu_6Sn_5 (thickness 5 –10 µm) has formed, **fig. 4(left)**. **Fig 4 (middle)** shows the joining zone of the alloy # 4 (ZnAl6Ag6CeGa) with the base material AlMg3. At the interface a structure with eutectic composition (Zn-Al) has formed.

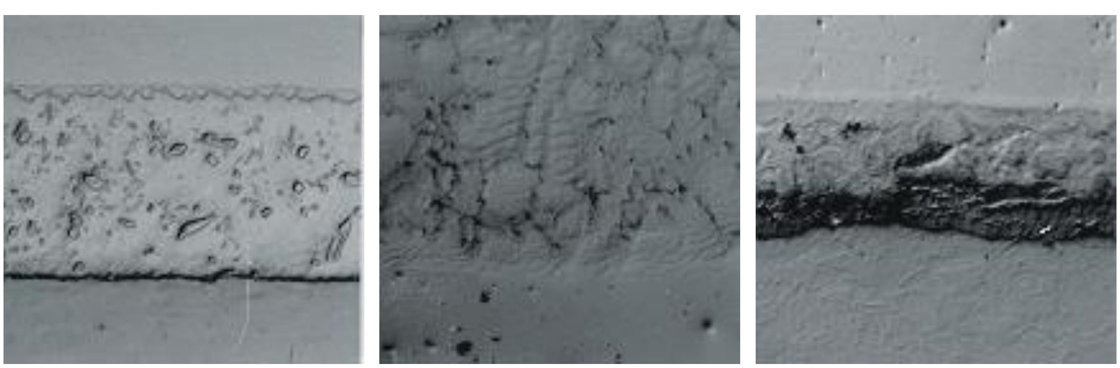

Fig. 4: E-Cu - sold.# 1- stainless steel (l), AlMg3-sold.# 4 (m), steel-sold.# 4-AlMg3 (r)

Mean shear strength values with AlMg3, E-Cu57 and steel ranged from 30 – 40 MPa. Maximum values of 57 MPa have been achieved on AlMg3, fig. 6 (left hand side). Tensile strength test with Al Cu2 Mg1,5 Ni (AA 2618) as base metal revealed tensile strength values over 100 MPa for all employed solders (# 1, 5, 6), **fig. 5** (right hand side). The maximum tensile strength is 138 MPa.

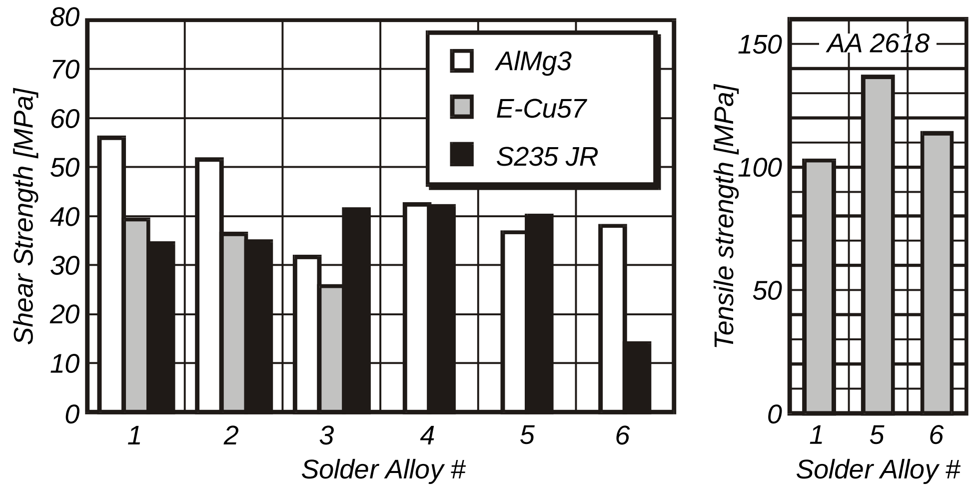

Fig. 5: Shear strength (left) and tensile strength (right) tests

APPLICATIONS

Heat Exchangers, Pipes and Fittings

Heat exchangers, pipes and fittings are typically made from aluminum, copper and steel. Conventional joining is done by brazing in protective atmosphere or under vacuum. Dissimilar materials require premetallization procedures. Gluing is otherwise used, which comes along with the disadvantage of low heat conductivity. With the presented technology similar and dissimilar materials can be joined. **Fig. 6 (left)** shows fins of AlMgSi0,5 soldered to a plate of AlMg3. Dissimilar materials joining is possible without prior metallization. Pipes from aluminum have been directly joined to copper tubes, **fig. 6 (right)**. Joining of aluminum to steel can be done without plating the steel part prior to the joining operation, **fig. 4 (right)**.

Fig. 6: Fins (AlMgSi0,5) - solder # 1 – plate (AlMg3) (left) and tubes (Al and Cu) (right)

Porous Materials

Fig. 7 demonstrates joints between valve bodies made from Ni-metallized brass and a porous steel filter. The decisive criterion for proper operation of the part is, that the pores of the filter metal don't get closed up by the filler metal. Due to the low capillary action of the employed solder (SnAg4Ti4CeGa) the liquid filler doesn't infiltrate the porous material.

Fig 7: Porous steel filter, soldered to Ni-metallized brass part

PVD-Targets

Targets being used in PVD-technology consist of a copper base plate and the target material to be deposited on substrate materials. Examples are chromium and alumina. The coefficient of thermal expansion (CTE) between the copper plate and the target material are significantly different. In that case low melting solders (e.g. SnAg4Ti4CeGa) are advantageous to keep induced residual stresses on a low level, **fig. 8**. Tests have demonstrated that the use of thermally sprayed intermediate layers are suitable when the PVD-process is run with enhanced wattages.

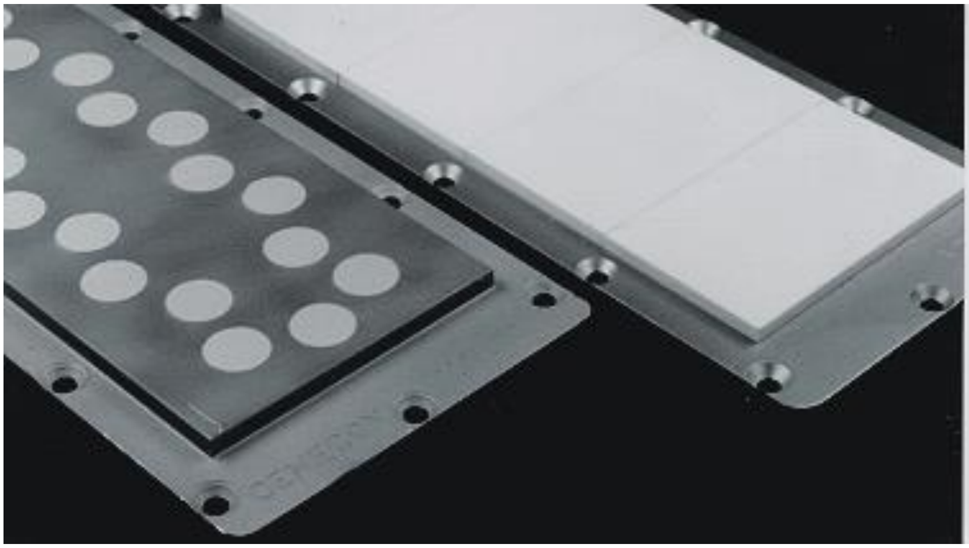

Fig 8: Titanaluminum (left) and alumina (right) soldered on copper plate

SUMMARY AND CONCLUSIONS

A system of tin and zinc based solders in conjunction with a appropriate process technology enables fluxless joining of materials like aluminum, copper and steel. Wetting of ceramics without premetallization is feasible. In order to produce ceramic-metal joints the problem of residual stresses due to mismatch of CTE of the parts to be joined has to be addressed as well. The use of thermally sprayed coatings is one solution to this problem. Soldering temperature ranges from 250°C (482°F) to 450°C (842°F). The removal of surface oxides has been done mechanically. Maximum shear strength is 57 MPa on AlMg3 and maximum tensile strength is 138 MPa for AA 2618 (Al Cu2 Mg1,5 Ni).

REFERENCES

(Ref. 1) Boretius, M; Lugscheider, E. and W. Tillmann.: VDI-Verlag, Düsseldorf, 1995 Joining of advanced ceramics

(Ref. 2) Lison, R.: DVS, Düsseldorf, 1998, Material joining by welding and soldering

(Ref. 3) Lugscheider, E.; Tillmann, W. and H. Zhuang: Patent# DE 19526822 C2, 1995 Solder alloy, use of solder alloy and method for joining workpieces by brazing

(Ref. 4) Rass, I., Lugscheider, E. and Hillen, F.: Patent application WO 99/02299, Solder braze alloy

(Ref. 5) Smith, R.W.; Lugscheider, E.; Hillen, F. and Rass, I.: Patent application# WO 99/12689, Process of using an active solder alloy

(Ref. 6) Lugscheider, E; Kortenbruck, G. and Scheer, K.: Schweißen und Schneiden 51 (1999), #5, P. 258 – 264, Manufacturing and evaluation of thin surface brazed coatings