JOINING OF COPPER-CARBON FIBRE COMPOSITE WITH GOLD COATED ALUMINA BY LOW TEMPERATURE In-Pb AND SOLDAMOL 170 SOLDERS

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In the paper the conditions of joining of copper matrix-carbon fibre composite with thin gold layer coated alumina by In50Pb50 and Sn60Pb36.5Ag3.5 (Soldamol 170) solders and their influence on the joint strength are presented. Joining was done in vacuum of the order of 10^{-3} Pa at the temperatures 483–523 (553) K during 600–1800 s. Joint strength (shear strength) was measured by push-off method in Instron testing machine. Maximum shear strength with Soldamol 170 solder, 23.4 MPa, was reached for joining temperature 523 K and time 600 s, maximum shear strength with In-Pb solder, 17.2 MPa, was reached for joining temperature 523 K and time for both solders until defect in joint integrity originates due to the difference in thermal expansion of ceramic, solder and composite. The joint structure study reveals that joint strength is controlled by the mutual diffusion of gold and indium into InPb solder, and gold and tin in Soldamol 170 solder, and by the development of the phases corresponding to AuIn₂ and AuSn₄, respectively.

 ${\rm K\,e\,y}$ w ord ${\rm s:}$ composite material, copper matrix, aluminium oxide, joining, joint strength

SPÁJANIE KOMPOZITU MEĎ-UHLÍKOVÉ VLÁKNO SO ZLATOM POKRYTÝM OXIDOM HLINÍKA NÍZKOTEPLOTNÝMI SPÁJKAMI In-Pb A SOLDAMOL 170

V práci uvádzame podmienky spájania kompozitného materiálu meď-uhlíkové vlákno s oxidom hliníka pokrytým tenkou vrstvou zlata spájkami In50Pb50 a Sn60Pb36,5-Ag3,5 (Soldamol 170) a ich vplyv na pevnosť spoja. Spájali sme vo vákuu rádu 10^{-3} Pa pri teplotách 483–523 (553) K počas 600–800 s. Pevnosť spoja sme merali na Instrone metódou push-off. Maximálnu pevnosť spoja so spájkou Soldamol 170 – 23,4 MPa – sme dosiahli pri teplote spájkovania 523 K a pri čase 600 s. Maximálnu pevnosť spoja

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so spájkou InPb – 17,2 MPa – sme dosiahli pri teplote 553 K a pri čase 1800 s. Pevnosť spoja pri obidvoch spájkach vzrastá s časom a s teplotou spájkovania až do vzniku poruchy integrity spoja v dôsledku rozdielu teplotnej rozťažnosti keramiky, spájky a kompozitu. Zo štúdia štruktúry spoja je zrejmé, že vzájomná difúzia zlata a india v InPb spájke a zlata a cínu v spájke Soldamol 170, ako aj vznik fáz so zodpovedajúcim zložením AuIn₂ a AuSn₄ vplývajú na pevnosť spoja.

1. Introduction

Alumina is frequently used in electrical and electronic industry as a carrier of electronic elements. High quality of these elements on a unit plane of a carrier and/or more powerful elements located on the carrier requires dissipation of generated heat. Heat sink material should possess the highest possible thermal conductivity and should be well joined with the ceramic carrier. Because of this requirement the coefficient of thermal expansion of both parts, ceramic and heat sink, should be very close. Another requirement for successful work of the heat sink is as low temperature of joining as possible to preserve the electronic elements to work properly.

Copper matrix-carbon fibre (Cu-CF) composite can be used for many applications (e.g. [1]). Very good properties of both components of the composite can be utilised for heat sink. These can be controlled by the choice of the carbon fibres amount in copper matrix, their arrangement and by the types of the fibres. Copper possesses high thermal conductivity (400 W/mK) and coefficient of thermal expansion of carbon fibre is negative. Except this some kinds of carbon fibres show also high thermal conductivity (e.g. Thornel K1100 shows ~ 1000 W/mK) and, thus, it is possible to produce the composite with relatively high thermal conductivity and thermal expansion close to that of alumina [2, 3].

Joining of metal matrix composites with metal, metal alloy or ceramics is more complicated than joining of common metals. Attention has to be paid to preserve the properties of the composites, i.e. the properties of reinforcing elements and their distribution in the matrix. There are many methods of joining of composites with metal or ceramics with the same or different composite. Ellis [4] gives a review about joining of aluminium based metal matrix composite. Woinski [5] presents general requirements for joining composite materials. The ideal process of bonding composites should proceed at the lowest temperature, within the shortest possible time and under the lowest possible pressure. Pietrzak et al. [6] studied the joining of nickel coated copper matrix-short carbon fibre composites with gold coated alumina ceramics by Sn-3Ag and Sn-3Cu solders. The best results were obtained with Sn--36.5Pb-3.5Ag solder. The average shear strength of the joining was around 25 MPa, which was obtained at the following joining parameters: temperature 483 K (210 °C), time 180 s, vacuum 1.3×10^{-3} Pa.

The aim of this paper is to study the processes of joining of copper matrixcontinuous carbon fibre composites with gold coated alumina ceramic by low temperature solders. The influence of joining parameters (temperature, time, type of solder) on the joining structure and bond strength was investigated.

2. Experimental procedure

Torayca T 300 carbon fibres containing 3000 monofilaments in a tow were chemically coated by copper followed by galvanic deposition of the copper to the thickness corresponding to 50 vol.% of copper in a composite [7]. Copper coated carbon fibres were woven into the unidirectional tapes. These were wafted by thin copper wire. The tapes were cut to the dimension 100×50 mm and put into pressing form with the fibre orientation 0 and 90° . Between the tapes, copper foils of 0.06 mm thickness were inserted. Number of the tapes gave the thickness of the specimen. Such Cu foils were given also on both outer sides of the specimen. The specimens for joining were produced by hot pressing of the tapes at the temperature of 873 K (600 $^{\circ}$ C) in vacuum of the order 10⁻³ Pa and cut to the dimensions $10 \times 10 \times 2$ mm. Alumina ceramic specimens coated by gold were cut to the dimension $12 \times 12 \times 0.6$ mm. Two solders were used: Foil of the In50Pb50 alloy of the thickness 0.05 mm with melting point $\sim 208 \,^{\circ}$ C made by rapid quenching in the Institute of Physics of the Slovak Academy of Sciences and alloy in the foil shape of Sn60Pb36.5Ag3.5 (Soldamol 170) from Degussa of the thickness 0.18 mm with the melting point 453 K ($\sim 180 \,^{\circ}$ C). Alumina was inserted into graphite holder and solder was put on the gold-coated side of the ceramic. Finally copper-carbon fibre composite was put on it. Four specimens were prepared for a given set of joining parameters: three were used for bond strength measurement and fourth was used for structure investigation by scanning electron microscopy equipped with X-ray analyser. Joint composition along the white line at 10 points is shown in Figs. 2-4. Shear strength was measured by Instron testing machine. Joining parameters were: temperature 483 K, 503 and 523 K (210, 230 and 250 °C), time 600, 1200 and 1800 s, and 553 K (280 °C), 1800 s and vacuum $\sim 10^{-3}$ Pa.

3. Results and discussion

a) InPb solder

In50Pb50 solder wets both to be joined materials (gold coated side of alumina as well as Cu-CF composite) well. Solder spilt on planes of both materials at 503 K (230 °C). Bond (shear) strength of the joint between gold coated alumina and Cu-CF composite formed by soldering with In50Pb50 for various joining temperatures and joining times is the Fig. 1.

Both parts (Cu-CF composite and gold coated alumina) for all joining times did not join at the temperature 483 K (210 °C). Temperature was too low to create sufficient diffusion. At 503 K (230 °C) the shear strength increases with joining time, for 523 K (250 °C) it is for all joining times approximately the same (~ 15 MPa) and

for 553 K (280 $^{\circ}$ C) shear strength (for 1800 s) decreased to 6.7 MPa. Structure of the joints is changing with temperature and joining time. Going from alumina



Fig. 1. Strength of the joint between alumina coated by gold and Cu-CF composite prepared with In50Pb50 solder for various joining temperatures and times.

side, a layer of gold can be seen both for low temperature and time (Figs. 2a,b), which at higher joining temperatures and/or times diffuses into solder, and indium diffuses into gold. The thickness of this diffusion zone increases with joining time at the given temperature while the thickness of the second part of the joint structure, which contains PbIn solder, decreases (Figs. 3a,b). For the temperature of 503 K ($230 \,^{\circ}$ C) and lower joining time (600 and 1200 s),PbIn solder is in contact with copper (as a part of composite), for higher time and higher temperature, the third zone is formed by the mutual diffusion of indium and copper (Figs. 3a,b). For the



Fig. 2. Structure of the joint made at 230 °C and 1200 s (a), relevant concentration profile of Au, In, Pb, Al, and Cu elements (b).



Fig. 3. Structure of the joint made at 250 $^{\circ}\mathrm{C}$ and 600 s (a) and relevant concentration profile (b).



Fig. 4. Structure of joint made at 280 $^{\circ}\mathrm{C}$ and 1800 s (a) and relevant concentration profile (b).

temperature of 553 K (280 $^{\circ}$ C) on both sides of the joint the phase of the composition arises close to AuIn₂ (Figs. 4a,b).

In all tested specimens the failure of the joint started in solder except in the case of joint made at the highest temperature 553 K (280 °C) where failure of the joint was at the composite and AuIn₂ phase interface, which can be a reason of the lower shear strength of the joint. From the concentration profile of all pictures it can be seen that gold and indium from InPb solder mutually diffuse and thickness of InPb solder decreases. Solubility of indium in gold is almost 10 at.% and solubility of lead in gold is practically zero. Solubility of indium in copper is up to 11 at.% while solubility of lead in copper is very low. From Fig. 4b it can be seen that mutual diffusion of gold and indium continues, and a phase corresponding to the composition AuIn₂ occurs. The same phase corresponding to AuIn₂ occurs on the other side of joint area (adjacent to composite).

b) Soldamol 170 (Sn60Pb36.5Ag3.5) solder

Soldamol 170 solder also wets well both parts of the materials to be joined. The strength of the joint made at 210 °C for 600 and 1200 s was practically zero. For 1800 s the strength was 8.8 MPa. This temperature (210 °C) is again too low to start sufficient diffusion between the solder and adjacent component (gold and copper). Joint strength for various joining time at the temperature 230 and 250 °C is in Fig. 5. The joint strength increases with joining time at 230 °C and maximum of the strength was reached at 250 °C for 600 s. The increasing of the strength is connected with the diffusion of gold into solder (Figs. 6a,b). From this picture it can be seen that the gold diffused across the whole solder forming a phase corresponding to the composition of AuSn₄. Higher joining time (at this temperature) gives rise to decrease of the strength due to high different coefficient of thermal expansion



Fig. 5. Strength of the joint between alumina coated by gold and Cu-CF composite prepared with Soldamol 170 solder at 230 and $250\,^\circ$ C for various joining time.



Fig. 6. Structure of the joint made at 250 °C and 600 s with Soldamol 170 solder (a) and relevant concentration profile (b).

between alumina and solder, and thus it comes to the failure of the integrity of the joint (Fig. 7).

Soldering does not involve the melting of the base metals so that problems of matrix reinforcement reaction, reinforcement diffusion, etc., can be reduced. Both studied solder materials were melted much below the melting point of the copper, even below the temperature $(600 \,^{\circ}\text{C})$ at which the voids start to originate in composite due to low adhesion between fibre and copper [8, 9]. From the experiments it can be seen that there is an optimum soldering temperature at which joint strength is maximum: below the temperature, joint fails in shear at the brazed interface (not enough developed diffusion), while at higher temperature the joint fails be-



Fig. 7. Structure of the joint made at $250 \,^{\circ}\text{C}$ and $1800 \,\text{s}$ with Soldamol 170 solder.

cause of the growing difference in the coefficient of thermal expansion of the components.

From the experiments it can be seen that both solder materials can be used for joining of alumina coated by gold with copper matrix-carbon fibre composite.

4. Conclusions

The obtained results can be summarized as follows:

1. Both low temperature Pb50In50 as well as Soldamol 170 solders wet well both parts of the joint: copper matrix-carbon fibre composite and gold coated alumina already at 483 K (210 °C).

2. Joint strength for both types of solders made at $210 \,^{\circ}$ C is low, even zero, due to not enough developed diffusion of gold into solder.

3. Optimal temperature of joining from the joint strength point of view is 523 K (250 $^{\circ}$ C).

4. Strength of the tested joints depends on the mutual diffusion of gold and indium in the case of PbIn solder and gold and tin in the case of Soldamol 170 solder.

5. Higher temperatures lead to bigger difference in thermal expansion between arising new phase in joint and ceramics, which leads to lower strength.

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