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RESEARCH ARTICLE

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Development of new transient liquid phase system Au-Sn-Au for microsystem technology

Abstract In the last decade, microsystems evolved to decisive technology in many technical applications. With increasing requirements on the performance of microsystems, more and more dissimilar materials are used in the same assembly. Correspondingly, suitable joining methods are required to fulfil the requirements on good properties of joints. In this study, a new transient liquid phase (TLP) system Au-Sn-Au was developed for potential medical applications in hybrid microsystems. The high and low melting phases Au and Sn were deposited onto diverse substrates by magnetron-sputter-ion plating. The coated substrates were soldered in a microsoldering station under different conditions. The influence of soldering conditions on the microstructure and properties of the joints was investigated. Results show that the developed solder led to high-quality joints that can be used in microsystems for medical applications.

Keywords transient liquid phase, microtechnology, soldering, diffusion, physical vapor deposition (PVD)

1 Introduction

In many technical fields, such as aerospace, medicine, optics, electronics, and biology, miniaturization has become one of the key technologies for decisive technical improvements. In hybrid microsystems, parts of dissimilar materials have to be joined. Soldering and brazing are often used to prepare joints with good mechanical and thermal properties. With respect to the special characteristics of microsystems, conventional solders usually lack the ability of wetting different types of substrate materials, especially materials like ceramics and glass. The small dimensions of the components to be joined complicate the positioning of the solders and their handling during the soldering process.

These technical challenges led to increased research activities to adapt transient liquid phase (TLP) solders to meet the special needs of microsystem technology. TLP-solders consist of a high and low melting phase. During the soldering process, the temperature is higher than the melting temperature of the low melting phase. Elements of the high-melting phase diffuse into the liquid and change the chemical composition, which leads to an isothermal solidification of the low melting phase. As a result, the remelting temperature of the joint is higher than the soldering temperature (see Fig. 1). The soldering temperature is mainly determined by the melting temperature of the low melting phase, which is usually composed of low melting elements like Indium (156.6°C), Tin (231.9°C), and Bismuth (271.2°C). Especially with eutectic compositions of these elements, the melting temperature can be reduced. The high-melting phase has the function to quickly diffuse to the liquid phase and to provide good adhesion properties on substrate materials. Typical high-melting materials are gold, copper, and nickel [1–3].

To assure a true to size application of solders onto the substrates in a submillimeter range and good adhesion of the high-melting phase on different substrates, physical vapor deposition (PVD) by magnetron-sputter-ion-plating (MSIP) can be used. The good adhesion results from the plasma cleaning of the substrate as well as the fact that the substrate surface does not have to be wetted in the soldering process. The geometric accurate application of the solder can be realized by suitable masking techniques [4,5].

In this study, the transient liquid phase system Au-Sn-Au was developed, primarily for potential medical applications in hybrid microsystems. The high and low melting phases Au and Sn were deposited onto diverse

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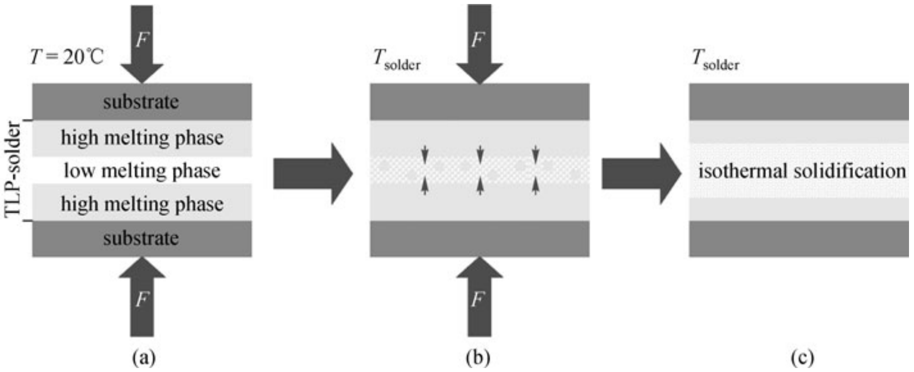

Fig. 1 Transient liquid phase process. (a) Initial solder composition; (b) solder at soldering temperature, diffusion of high-melting elements to liquid phase; (c) soldered joint after isothermal solidification

substrates by magnetron-sputter-ion plating. The coated substrates were soldered in a microsoldering station under different conditions. The influence of the soldering conditions on the microstructure and properties of the joints was investigated. The results showed that the developed TLP solder can realize high-quality joints to be used in microsystems for medical applications.

2 Experimental

Au and Sn were consequently deposited onto diverse substrates by magnetron-sputter-ion plating using a Leybold LH Z400 device. The deposition parameters are listed in Table 1. To increase the adhesion on substrates, a very thin Ti layer (−200 nm) was deposited first. Two coated substrates were used to produce a joint in a microsoldering station. The soldering parameters are given in Table 2. The soldered joints were metallographically prepared and characterized in terms of their microstructure using scanning electron microscopy (SEM) equipped with Energy dispersive X-ray spectroscopy (EDS).

To determine the mechanical properties of the joints, shear tests [6] were carried out with the substrate combinations of silicon/borosilicate glass, AlN/GaAs, Cu/AlN, and silicon/Al_2O_3 based on the standard DIN EN 12797. The thermo-shock resistance of the joints was evaluated by dipping the joints alternately into oil bathes of 180°C and 20°C, each for 45 s. This procedure was repeated up to 100 times. The number of cycles was noted until a joint was macroscopically damaged.

The corrosion resistance of the soldered joint was evaluated regarding the industrial standard DIN 50905 by deposition of the joint in a corrosion testing device using a simulated body fluid (SBF: Ion concentration: 142 mM Na^+, 5 mM K^+, 1.5 mM Mg^{2+}, 2.5 mM Ca^{2+}, 148.8 mM Cl^-, 4.2 mM HCO_3^-, 1 mM HPO_4^{2-}, and 0.5 mM SO_4^{2-}) at 37°C for four days. The SBF was nebulized continuously in the device, so the joint was permanently covered with a thin SBF film. After the immersion test, the mass change was determined, and the microstructure was investigated by scanning electron microscopy.

3 Results and discussion

The deposition trials by magnetron-sputter-ion plating show that the high-melting phase Au can be well deposited onto diverse substrates under the given deposition conditions. The low melting phase Sn can be well deposited on the predeposited Au layer. The thickness of the single layer can be exactly controlled by the deposition parameters. For instance, Fig. 2 shows a cross-section of Au-Sn on glass and Fig. 3 the calotte projection. In the calotte projection, the thin Ti-layer is also visible. It is evident that the Au layer adheres very well to the substrates. No defects are visible at the interfaces of the layer to the devise substrates. The Sn layer also shows a good adhesion to the Au layer. Figure 2 shows the sharp interface between the Sn and Au layers indicating a high purity of the faces even in the bounding surface. This indicates that the chemical composition of the solder system can be exactly controlled by properly selecting the coating thickness.

The soldering trials show that all coated substrates can be soldered well under the given soldering conditions. For example, Figs. 4–6 show some soldered joints with different substrate combinations. It is evident that all the

Table 1 Parameters for MSIP process

	pressure/Pa	power/W	substrate temperature/°C	pulse/kHz	cooling break/min
Au-Sn	2–0.5	250–100	−35/5	50	15–30

Table 2 Soldering parameters

soldering temperature/°C	duration/min	compression/MPa
27–400	60	0.5

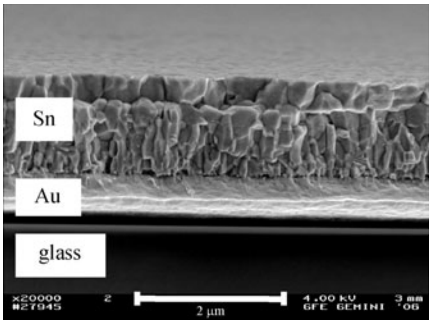

Fig. 2 Cross-section of an Au-Sn-Ti coated glass substrate

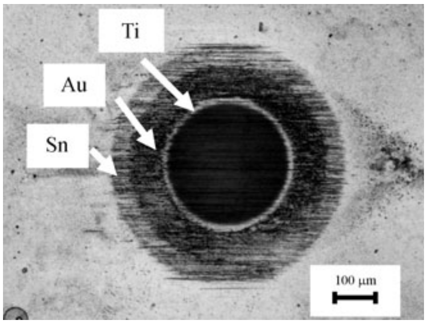

Fig. 3 Calotte projection of an Au-Sn-Ti coated glass substrate

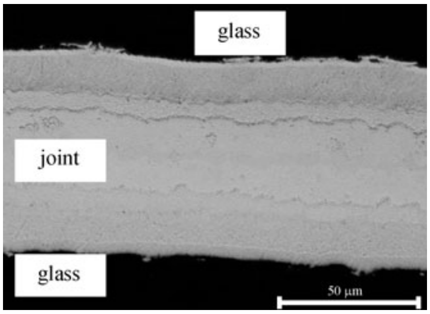

Fig. 4 Cross-section of joint with glass substrates soldered at 270°C for 30 min

soldered joints show a perfect connection of the solder seams to the substrates due to the predeposition by magnetron-sputter-ion plating. However, the microstructure of the joints varies significantly depending on the soldering conditions and the solder compositions.

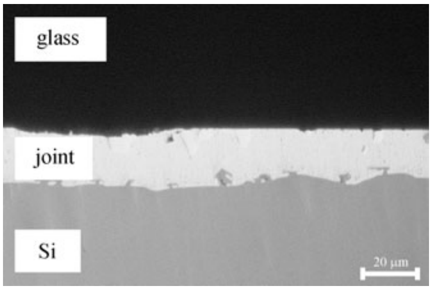

Fig. 5 Cross-section of joint with glass and Si substrates soldered at 400°C for 1 h

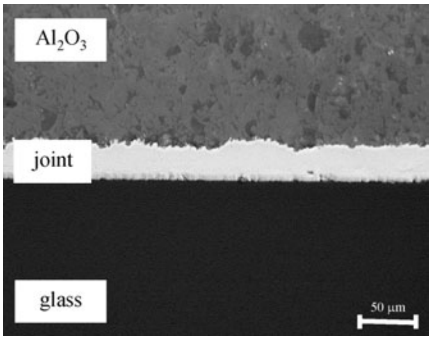

Fig. 6 Cross-section of joint with Al_2O_3 and glass substrates soldered at 400°C for 1 h

The influence of the soldering conditions on the microstructure of the soldered joint was deeply investigated using a coating thickness proportion of 10:3 (Au:Sn). Corresponding to this proportion, the complete coating has a Sn concentration of about 15 at. %. Figure 7 shows cross-sections of the joints soldered at 270°C for 60 min. and at 400°C for 60 min, respectively. Both the joints show no defects in the solder seams. The solder seam shows a perfect connection to the substrates. However, the microstructures of the both joints differ significantly. The joint soldered at 270°C shows a multilayer microstructure, whereas the joint soldered at 400°C shows a homogenous microstructure. EDS measurements show that the layers marked with No. 1, 2, and 3 mainly consist of AuSn, $AuSn_2$, and $AuSn_4$. The formation of the different microstructures can be explained with the aid of the phase diagram Au-Sn (Fig. 8). When the temperature reaches the melting point of Sn at 232°C during heating, the Sn layers on the both substrates melt and fuse together to a single liquid layer in the middle of the both substrates. After melting of Sn layers, the Au layers begin to dissolve into the liquid layer. The dissolution of the Au layer continues, and the Au diffuses into the mid of the liquid layer from the both interfaces of the Au layers. A gradient in the Au concentration forms in the liquid layer. The Au concentration decreases from the interfaces to the middle of the liquid layer. The dissolution of the solid layers will

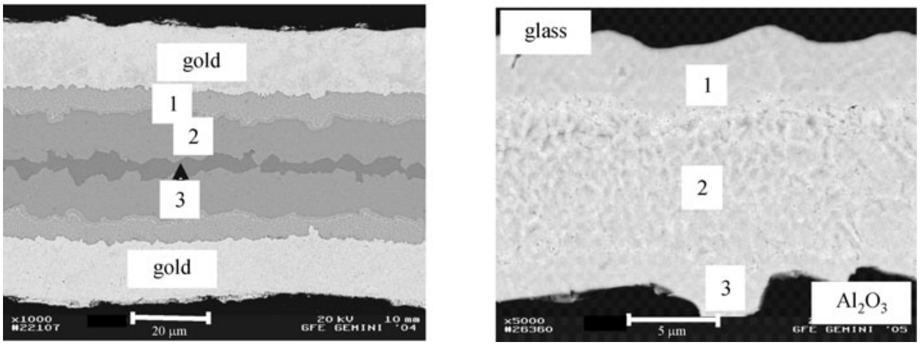

Fig. 7 Left: soldered at 270°C for 60 min with resulting phases AuSn-δ(1), $AuSn_2$-ε(2), and $AuSn_4$-η(3); right: soldered at 400°C for 60 min with homogeneous gold rich phase (more than 10 at.-% Sn) at (1), (2), and (3)

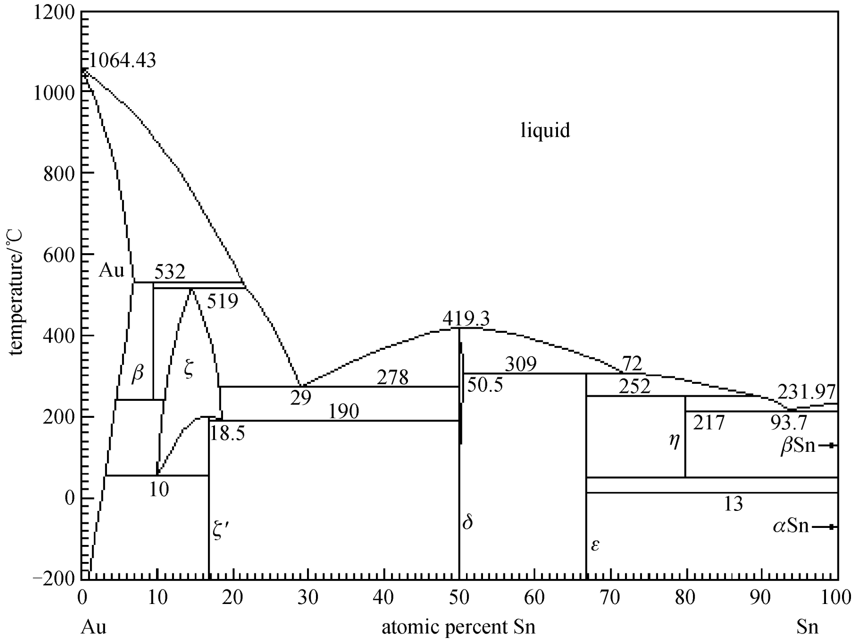

Fig. 8 Phase diagram of Au-Sn

stop when the Au concentration in the liquid layer near the interfaces reaches 15 at. %, at which the liquidus temperature is 270°C. From this time point, the new intermetallic phase $AuSn_2$ begins to solidify out of the liquid phase isothermally at the interfaces. Because of the gradients in Sn and Au concentrations, Sn further diffuses in the direction of the substrates, and Au further diffuses in the direction of the liquid layer. As a result of the diffusion processes, more and more liquid phases transform to $AuSn_2$ (ε-phase), and the liquid layer reduces continuously. Because of the continually diffusion of Au and Sn, the solidified $AuSn_2$ and Au layer near their beginning interfaces are not stable, and they transform to the new intermetallic phase AuSn (δ-phase). With increasing time, the thickness of the AuSn layer increases. The narrow $AuSn_4$ layer in Fig. 7 indicates that after the temperature begins to decrease, there is still a narrow liquid layer in the middle of the joint. During cooling to room temperature, no equilibrium reaction occurs from the remaining liquid, resulting in the formation of the metastable phase $AuSn_4$.

When the soldering process is repeated with a soldering temperature of 400°C, the solidification differs significantly from the abovementioned process. As shown in the phase diagram, the dissolution of the Au layers into the liquid layer can only stop when the Au concentration in the liquid layer reaches 74 at. %, at which the liquidus temperature is 400°C. After this concentration has reached, Sn and Au diffuse further, resulting in that the metastable

Table 3 Shear strengths of soldered joints with different substrates

	silicon/glass	AlN/GaAs	copper/AlN	silicon/Al_2O_3	steel/Al_2O_3
shear strength/MPa	42±2.4	45±2.8	46±2.8	44±2.6	58±3.4

ζ-phase begins to solidify at the interfaces. Because the thickness proportion of 10:3 corresponds to about 15 at. % Sn, the liquid layer can disappear completely with increasing time. After the liquid layer disappeared, the diffusion processes still takes place. As a result, the distribution of Au and Sn in the total joint homogenizes further, thus the total joint consists of the ζ-phase and Au solid solution at the soldering temperature. After the temperature decreases to 190°C during cooling, the ζ`-phase begins to precipitate, whereas the Sn concentration in the remaining ζ-phase reduces to 10 at. %. Finally, the microstructure consists of primary ξ`-phase and either a eutectic Au-ξ`-phase or a metastable ξ phase at room temperature. The results show that the joint soldered at 400°C does not contain the tin rich phases $AuSn_4$, $AuSn_2$, and AuSn, which have a lesser biocompatibility than the Au-rich phases. Based on these results, the joints were prepared with the coating thickness proportion of 10:3 (Au:Sn) at 400°C for 1 h for further investigation.

The shear tests show that all tested joints have high shear strengths in the band of 40–60 MPa (Table 3). The lowest value is 42 MPa for the Si/Glass combination. The highest value is 58 MPa for the steel 1.4401/Al_2O_3 combination. The higher strengths of the substrates of steel 1.4401 and Al_2O3 could be the reason for the highest shear strength. The results of the thermo-shock tests are presented in Table 4. All joints with the different substrate combinations but steel 1.4401/Al_2O_3 showed no defects after 100 cycles. Only the joint with the substrate combination steel 1.4401/Al_2O_3 failed after 87 cycles. The higher strengths of the substrates and large difference in their thermal expansion coefficients should be the reason for the failure, because they could cause higher stresses in the joint area during thermo-shock test.

Table 4 Number of thermo-shock cycles without the occurrence of macroscopical damages

substrate materials	cycles
copper/silicon	100
silicon/glass (pyrex)	100
Al_2O_3/steel (1.4401)	87
copper/AlN	100
AlN/Silicon	100
GaAs/AlN	100

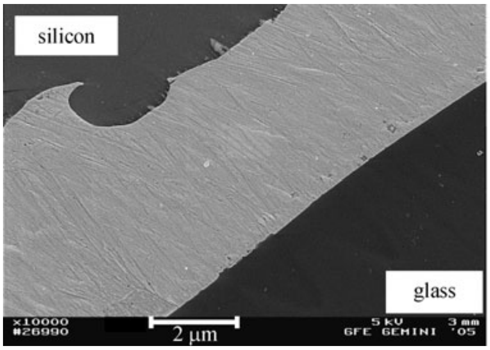

Fig. 9 Soldered joint after four days in simulated body fluid

The corrosion test shows that the soldered joint has a high corrosion resistance under the test conditions. No weight loss could be determined, and no signs of a corrosive attack in the soldered joint are visible in the SEM micrograph of the joint (Fig. 9), indicating that the developed Au-Sn-Au system is suitable for medical applications.

4 Conclusions

The transient liquid phase solder Au-Sn-Au was developed and characterized in this study, primarily for potential applications in the medical area. The high and low melting phases Au and Sn can be well deposited on different substrates by magnetron-sputter-ion plating. The coated substrates can be well joined under different conditions. The joints prepared with the optimized compositions and soldering parameters show homogenous microstructures, high shear strengths, good thermo-shock behavior, and high corrosion resistance and can be used in hybrid microsystems for medical applications.

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