

P. SKRZYNIARZ\*, A. SYPIEŃ\*, J. WOJEWODA-BUDKA\*, R. FILIPEK\*\*, P. ZIEBA\*

## MICROSTRUCTURE AND KINETICS OF INTERMETALLIC PHASES GROWTH IN Ag/Sn/Ag JOINT OBTAINED AS THE RESULTS OF DIFFUSION SOLDERING

## MIKROSTRUKTURA I KINETYKA WZROSTU FAZ W POŁĄCZENIU Ag/Sn/Ag UZYSKANYM W WYNIKU LUTOWANIA DYFUZYJNEGO NISKOTEMPERATUROWEGO

Copper plays the role of the fundamental conductive layer in electronic circuits. Nevertheless, in the crucial areas, silver is used due to its higher electrical and thermal conductivity as well as oxidation resistance. Diffusion soldering is one of the novel technologies of metals joining which can successfully be used in the electronic industry. The main advantage of this process is that a joint produced is characterized by thermal and mechanical stability at temperatures 2-3 times higher than the soldering temperature.

This paper presents results of optical and scanning electron microscopy observations of Ag/Sn/Ag joints obtained using diffusion soldering at the temperature 243 °C and 258 °C. The kinetics of Ag<sub>3</sub>Sn intermetallic phase growth was also studied as well as the chemical composition at the joint cross sections was determined using the energy dispersive X-ray spectroscopy method.

The interdiffusion coefficients were calculated on the basis of the determined Ag<sub>3</sub>Sn phase growth rate curves applying the numerical diffusion model developed at the Department of Solid State Chemistry, the Faculty of Materials Science and Ceramics at the AGH University of Science and Technology.

*Keywords:* diffusion soldering; microstructure; kinetics

Rolę podstawowej warstwy przewodzącej w układach drukowanych spełnia miedź. Mimo to w obszarach newralgicznych, stosuje się srebro ze względu na wyższą przewodność elektryczną i cieplną, a także odporność na utlenianie. Lutowanie dyfuzyjne niskotemperaturowe (*ang. diffusion soldering*) stanowi jedną z nowatorskich technik łączenia metali ze względu na łatwość zastosowania i szybkość procesu, w szczególności w przemyśle elektronicznym. Główną zaletą lutowania dyfuzyjnego niskotemperaturowego jest otrzymanie połączenia wykazującego stabilność termiczną i mechaniczną w temperaturach 2-3 razy wyższych niż temperatura lutowania.

W pracy przedstawiono wyniki badań złącz Ag/Sn/Ag otrzymanych za pomocą lutowania dyfuzyjnego niskotemperaturowego w temperaturze 243 °C i 258 °C wykonane techniką mikroskopii optycznej i skaningowej mikroskopii elektronowej. Określona została także kinetyka wzrostu fazy międzymetalicznej Ag<sub>3</sub>Sn i skład chemiczny na przekroju spoiny metodą spektroskopii promieniowania rentgenowskiego z dyspersją energii.

W oparciu o wyznaczone krzywe szybkości wzrostu fazy Ag<sub>3</sub>Sn obliczono współczynniki dyfuzji wzajemnej przy zastosowaniu numerycznego modelu dyfuzji opracowanego w Katedrze Fizykochemii Ciała Stałego na Wydziale Inżynierii Materiałowej i Ceramiki Akademii Górniczo-Hutniczej.

### 1. Introduction

Environment protection and improving the quality of joints are the two main bases of the present development of modern technologies of different materials joining. The environment protection trend began in the 1990's in the USA and later it also spread over Europe and Japan. The main aim is working out soldering materials

able to replace the Sn-Pb solders commonly used so far. This can be obtained by eliminating cadmium and lead which are the components of soft solder used in conventional soldering process. Additionally, the EU applied directive RoHS 2002/95/EC of January 27, 2003 orders the member countries to limit the use of some hazardous substances in electrical and electronic equipment. Soon, another directive was issued: WEEE 2002/96/EC

\* INSTITUTE OF METALLURGY AND MATERIALS SCIENCE OF POLISH ACADEMY OF SCIENCE, 25 REYMONTA ST., 30-059 KRAKÓW, POLAND

\*\* DEPARTMENT OF SOLID STATE CHEMISTRY, FACULTY OF MATERIALS 30-059 KRAKÓW SCIENCE AND CERAMICS, AGH UNIVERSITY OF SCIENCE AND TECHNOLOGY, 30 MICKIEWICZA AV., POLAND

– Waste Electrical and Electronic Equipment referring to the problem of the used up electrical and electronic equipment and its reusing, recycling, and other forms of recovery. It imposes the responsibility for storing and recycling hazardous substances on the manufactures.

Hence, a lot of information concerning new methods of joining materials can be found in the literature. The electronic industry is a good example. Assembly line production of circuits of high integration scale with many units sensitive to high temperature and joined in a very short time enforces applying a special soldering process so that the solder area is as small as possible. Diffusion soldering meets such requirements. The joint made in this way takes up to 6 times less space than in the case of conventional soldering and it can work at the temperatures higher than 350 °C [1, 2], and it often shows mechanical and thermal stability at temperatures 2-3 times higher than the joining temperature. Moreover, the surfaces to be joined do not require special preparation, which remarkably shortens the production time [3]. The example here could be circuits on the basis of SiC and semiconductors from the groups II-V, e.g. jet engines, nuclear reactors, industrial robots, car electronics, spaceships and satellites as well as assemblies of the metal-ceramics type. Besides, joints obtained using diffusion soldering work well in the situation in which high heat dissipation makes conventional soldering impossible to use, e. g. in the case of heating plate in electric cookers or different electronic elements (resistors or transistors) [4].

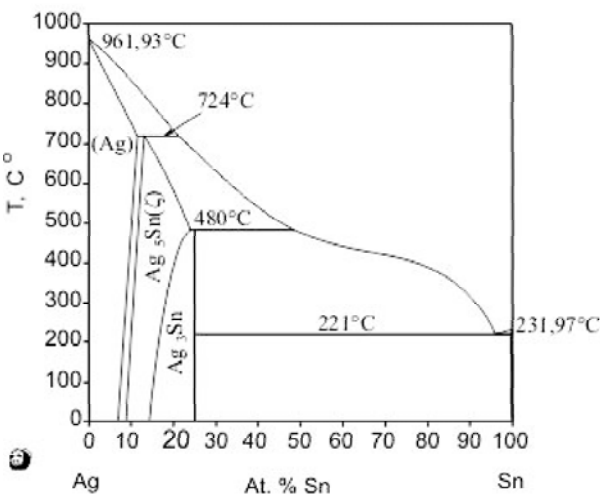


Fig. 1. Equilibrium phase diagram Ag-Sn (created in program Pandat) [5]

The appropriate equilibrium phase diagram is the basis for applying the diffusion soldering process (DS) because the aim of DS is obtaining an intermetallic phase or phases (IP) which fill up the whole joint area. Liquid phase is the transitional stage after which isothermal so-

lidification follows and then IP growth stage. Hence, it is very important to match an appropriate joining metal depending on its melting temperature, occurrence and thermal features of the obtained IP as well as their obtaining sequence which can be provided referring to the appropriate equilibrium phase diagram.

In spite that Cu plays the main role of the conductive layer in circuits, in crucial areas and joints silver is used because of its higher electrical (Cu-58; Ag-63 ( $\times 10^6$  S/m)) and heat conductivity (Ag-418; Cu-394 (W/(m\*K))) as well as oxidation resistance.

Jacobson and Humpston [6] were the first to examine the Ag-Sn system (Fig. 1). The formation of high-melting IPs of a small molar volume difference are its greatest advantages, which prevents the creation of voids during the solder solidification. During the isothermal annealing of the Ag/Sn/Ag assembly at 250 °C two IPs are obtained in the following sequence: Ag<sub>3</sub>Sn of the melting temperature 480 °C and Ag<sub>5</sub>Sn (724 °C). Applying a respectively thin layer of tin makes its entire conversion possible in a few minutes' time. Further heating leads to broadening of the reaction area due to the tin to silver diffusion and the appearance of the solid solution of tin in silver (Ag). Further experiments on this system were connected with working out a method of thyristors joining to molybdenum radiators [6]. Aluminum-silicon braze alloys used up to this moment together with the increasing diameter of devices were not resistant enough to mechanical stresses resulting from big differences in thermal expansion of silicon and molybdenum. The optimal heat output was obtained by applying appropriately thin and well filled up with solder the joints. Despite the fact that the Ag<sub>3</sub>Sn phase was present in the joint, still the unsoldering temperature was about 500°C and it grew up even to 750 °C together with enriching the joint center with silver [7]. After about 100 hours of annealing a solid solution of tin in silver (Ag) appeared [6].

In electronic industry the majority of circuits are build as the so called sandwiches which have specified tasks to carry out. The metals which are deposited in the form of a single layer or sandwiches are Au, Ag, Cr, Cu, Ni, Sn and Zn [8]. An example of a circuit may consist of the following sequence of layers: Si (SiO<sub>2</sub>)/Ti (Cr, Co, Co-P)/Cu/Ni/Ag (Au)/Sn (In, Sn-In) [9]. These coatings play the following functions [10]: Si (SiO<sub>2</sub>) is semiconductor and is the main carrying framework, the Ti layer or interchangeably Cr, Co or Co-P are the diffusion barrier, mainly electrodiffusion barrier for copper, Cu is the conductive base, Ni protects copper against oxidation, Ag or Au is a film of the highest conductivity, and Sn is deposited optionally as another shell protecting Au against oxidation and other reactions. Moreover, the Sn, In or Sn-In layers deposited only locally make a solder

for obtaining a joint e. g. using the diffusion soldering method [11].

As can be seen the application of silver as the conductive layer as well as the requirements of the end user referring to joints reliability and their durability and low environmental hazard are a good reason for detailed research of microstructure and chemical composition changes at the cross sections of the obtained Ag/Sn/Ag joints, which are an alternative to other types of joints.

2. Experimental

The samples for the investigation of microstructure and kinetics were prepared using the Sn foil of 99.9 % purity and of 0.127 mm thickness as a solder. The silver substrates were of 99.99 % purity and of 0.7 mm thickness cut out from a silver rod of Ø 7 mm employing a Well 3242 diamond wire saw using a diamond wire of Ø 3 mm diameter. The substrates surface was prepared by grinding using 180 - 1200 µm abrasive papers and

polishing with the use of 1 - 0.25 µm diamond paste. In the process of preparing the Ag substrates care was taken to obtain parallel of both sides of the substrate in order to get a joint of the same width on the joint cross section in the diffusion soldering process (DS). Next, the Ag substrates were rinsed with water and cleaned with acetone in the ultrasonic washer for 40 minutes, and the Sn foil for 20 seconds. The Sn foil was placed between two Ag substrates together with distancing tantalum strip and clamped in a press holder at the pressure of 3-4 MPa (Fig. 2a). The Ag/Sn/Ag pieces assembled and pressed in this way were put into the DS apparatus in order to join them at the constant temperature for selected time (Table 1).

TABLE 1  
Production conditions of diffusion joints of the Ag/Sn/Ag type

Temperature [°C]	Time [min]
243	10, 20, 30, 40, 60, 80
258	20, 30, 40, 50, 60, 80

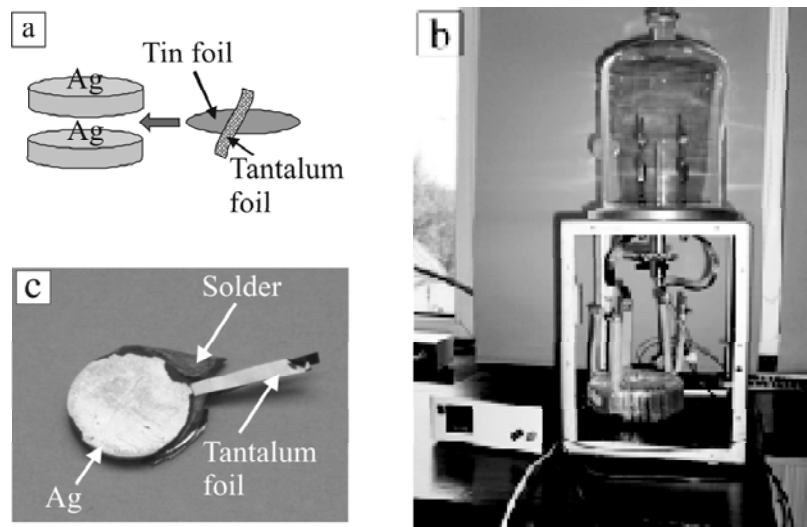


Fig. 2. Preparation of the diffusion soldered joints: (a) sample preparation diagram; (b) special furnace for DS; (c) example of a joint after DS

The Ag/Sn/Ag joints in unetched state were viewed on light microscope (LM) of the type Leica DMIRM at magnifications up to 500 times. The observations were carried out in the polarized light in order to obtain a preliminary information about the type of phases formed in the solder as well as to obtain a clear boundary outline of the appearing phases needed for further kinetics calculations.

The studies with the use of FEI scanning electron

microscope (SEM) E-SEM XL30 enabled detailed observations of the joint morphology, its quality, and evaluation of the number of phases, the width of which was too small to identify them in the process of the LM studies. The information about qualitative composition changes in the joint area was provided by the observations in the backscattered electrons mode (BSE). Chemical analysis were carried out using an Energy Dispersive X-ray Spectrometer (EDX) Link ISIS. Quantitative analysis with

ZAF correction method was performed with resulting relative error of 4 %.

3. Research results and discussion

3.1. Microstructure

The IP formation was initiated by the diffusion processes and dissolution taking place on the liquid/solid phase boundary between the melted tin and the silver substrate. The microstructure after the DS process at the temperature of 243 °C (Fig. 3) shows the presence of the IP scallops growing into the tin solder.

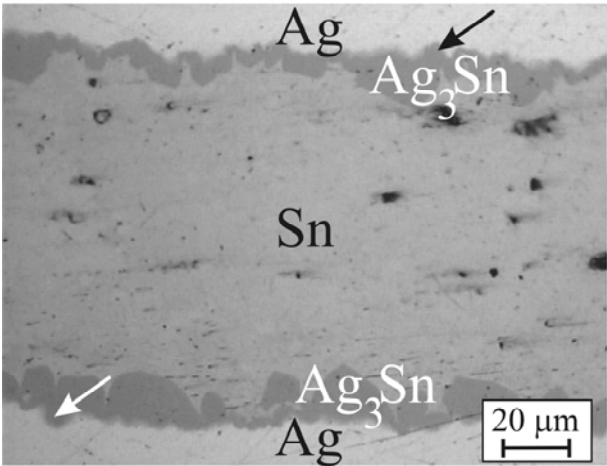


Fig. 3. LM image of DS joint Ag/Sn/Ag after annealing at 243 °C for 80 min.

Along with prolonging of the annealing time the IP is thicker and the smaller scallops, because of the Ostwald ripening process [12], get joined forming bigger ones separated by channels that are fast diffusion paths for liquid tin. The scallops are also occasionally formed on the silver side (Fig. 3 – see arrows) and a corrugated shape is visible on the boundary of the Ag/IP which indicates the fast dissolution and diffusion of Ag in liquid Sn [13].

For the joints obtained at the temperature of 243 °C (Fig. 3) the linescan of the chemical composition were carried out on the cross section of the Ag/Sn/Ag joint which confirmed formation of only one IP-Ag<sub>3</sub>Sn (Fig. 4).

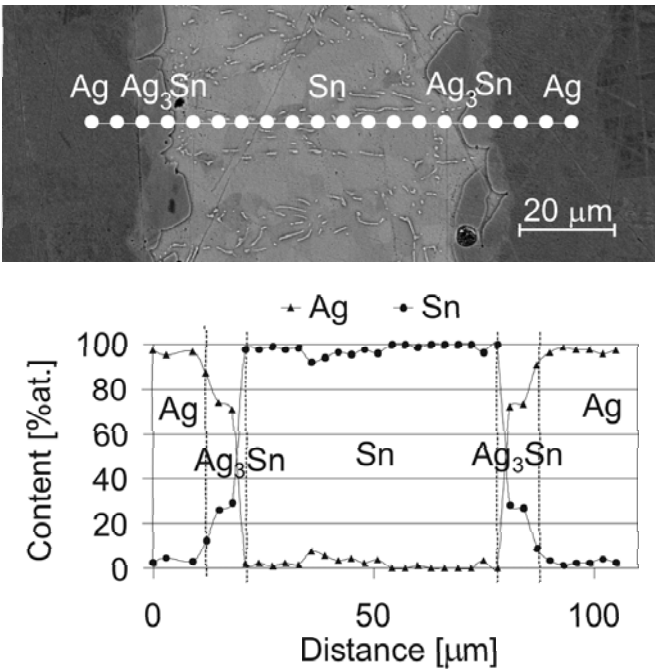


Fig. 4. SEM-BSE image and EDX linescan with 3 μm step Ag/Sn/Ag joint annealed at 243 °C for 20 min

The formation of Kirkendall voids (Fig. 5) from the side of the liquid solder confirms the difference in the diffusion rate of Ag and Sn through the Ag<sub>3</sub>Sn phase.

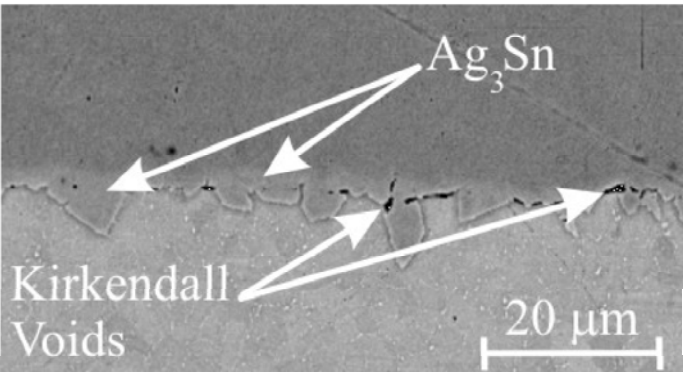


Fig. 5. SEM-BSE image of the Ag/Sn/Ag joint after annealing at the temperature 243 °C for 10 min

At the temperature of 258 °C, the formation of whiskers growing directly from the Ag substrate into of the tin was noticed after short annealing times (Fig. 6a – see arrow). In this case the whiskers growth may be caused by the formation of a layer of impurities on Ag or a remarkably dense and thick oxide coating on the Sn surface (Fig. 6a). The whiskers disappeared locally after penetration by liquid Sn. The growth of whiskers is attributed to two effects: a significant dissolution rate of Ag in Sn [13], a local supersaturation of the Ag-Sn solution by the dissolved Ag leading to the formation of Ag<sub>3</sub>Sn phase as a result of eutectic reaction during solidification [14, 15].

Similarly as for the temperature of 243 °C, at 258 °C the channels between IP scallops are formed and a corrugated shape is visible on the boundary of Ag/IP phases. However, after a longer annealing period the tin in the channels is enriched in Ag and a more uniform layer is formed (Fig. 6b).

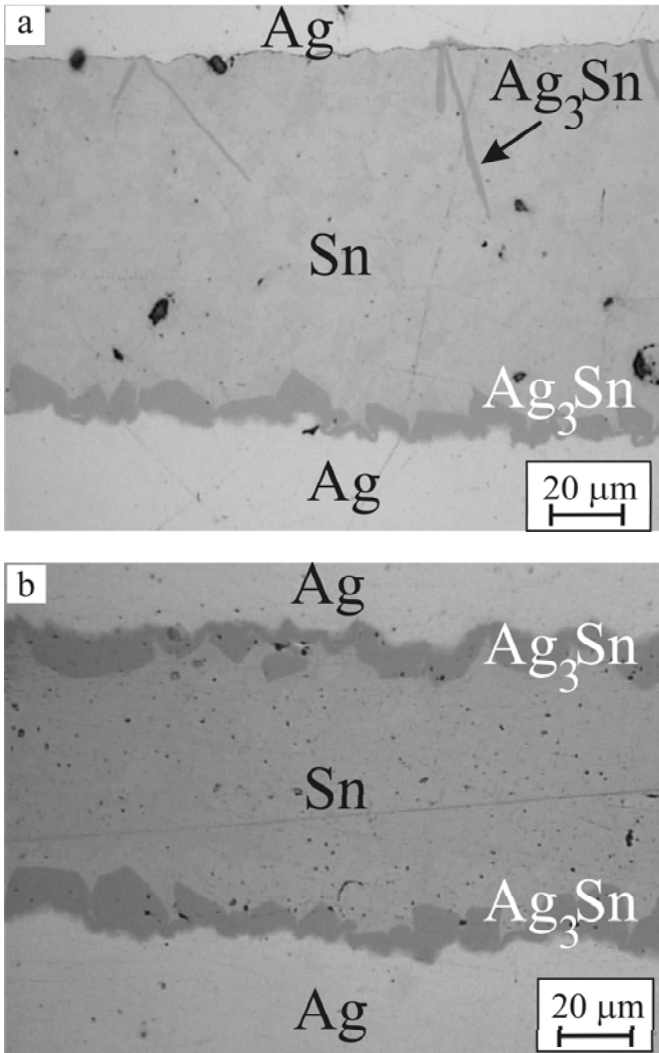


Fig. 6. LM images of DS joints Ag/Sn/Ag after annealing at 258 °C for 20 min. (a) and 50 min. (b)

Moreover, it was found that the parts being joined with a deposited layer of solder on them do not require a special treatment as the thin oxide coating that formed on Sn does not make a significant diffusion barrier.

For the joints obtained at the temperature of 258 °C a quantitative microchemical analysis was performed at the selected areas on the joint cross section, which confirmed that the growing IP corresponds to the composition of the  $\text{Ag}_3\text{Sn}$  phase i. e. Ag 74.9 % at. and Sn 25.1% at., and also in addition to that chemical analysis linescan was performed on the Ag/Sn/Ag joint cross section (Fig. 7).

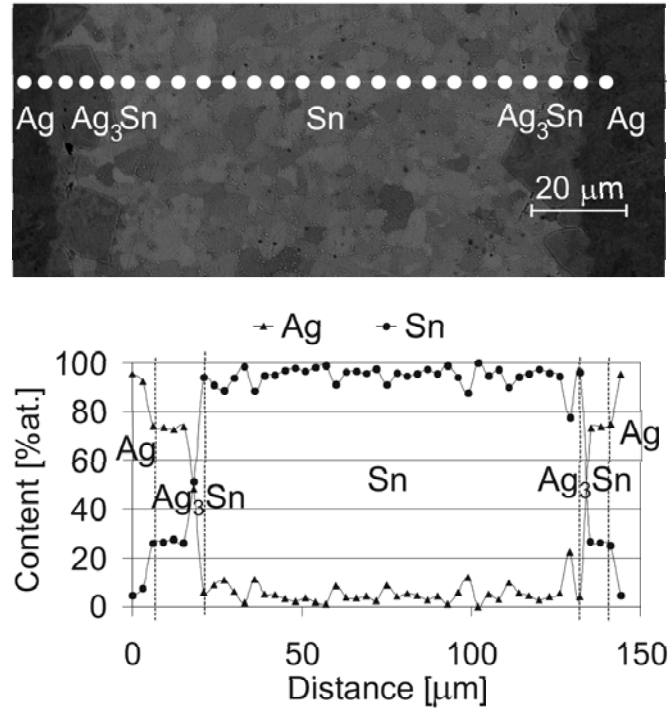


Fig. 7. SEM-BSE image and EDX linescan with 3  $\mu\text{m}$  step Ag/Sn/Ag joint annealed at 258 °C for 20 min

### 3.2. Kinetics

The examination was carried out for the temperatures of 243 °C and 258 °C. The following formula was used to describe the kinetics

$$d = k \cdot t^n \quad (1)$$

The exponential factors calculated for the temperatures 243 °C and 258 °C are 0.38 and 0.45, respectively. If the values of the exponential factor are below  $n=0.5$ , this corresponds to the volume diffusion, while for range  $n<0.5$  one can expect some contribution from the grain boundary diffusion.

Figures 8 and 9 present the growth rate curves in the form of relations:  $\text{Ag}_3\text{Sn}$  phase thickness  $d(x)$  versus the annealing time  $t$  and  $\log d$  versus  $\log t$  on the basis of which the values of  $n$  and  $k$  were determined by fitting procedure.

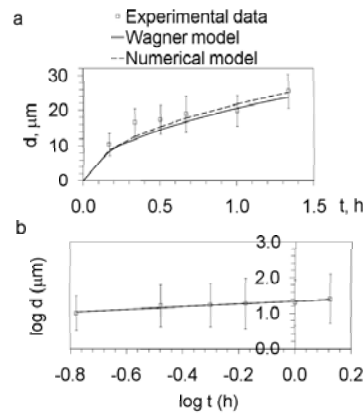


Fig. 8. The graph of Ag<sub>3</sub>Sn phase thickness vs. growth time (squares) together with the Wagner model (solid line) and the numerical model [16] (dashed line) for the Ag/Sn/Ag joints at the temperature of 243 °C in diagram: (a)  $d=f(t)$  and (b)  $\log d=f(\log t)$

According Van Loo and Pierragi [17, 18] classification, the shape curves show predominance a kind of diffusion. The results obtained in the first stage of the Ag<sub>3</sub>Sn phase growth, where relationship  $d$  versus  $t$  was linear and slope was the greater, indicate predominance the grain boundary diffusion (Fig. 8a). In the second stage, the parabolic shape of the graph shows the process stabilization and a possible the volume diffusion

predominates [17, 18]. Similarly, for the temperature of 258 °C (Fig. 9a) the first range indicates a higher rate of the Ag<sub>3</sub>Sn phase growth where the grain boundary diffusion predominates. The second range shows the stabilization of the Ag<sub>3</sub>Sn phase growth process and as rate controlling factor the volume diffusion [17, 18].

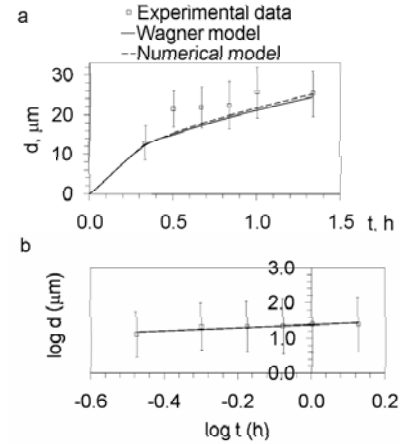


Fig. 9. The graph of Ag<sub>3</sub>Sn phase thickness vs. growth time (squares) together with the Wagner model (solid line) and the numerical model [16] (dashed line) for the Ag/Sn/Ag joints at the temperature of 258 °C in diagram: (a)  $d=f(t)$  and (b)  $\log d=f(\log t)$

TABLE 2  
The values of the  $n$  factor and the growth rate constant  $k$  of the Ag<sub>3</sub>Sn phase for different temperatures in the Ag/Sn/Ag joint

Temperature [°C]	$n$	$k$ [ $\mu\text{m}/\text{h}^n$ ]	Reference
230	$0,67 \pm 0,10$	$15,25 \pm 0,36$	[19]
235	$0,63 \pm 0,08$	$16,01 \pm 1,32$	[16]*
243	$0,38 \pm 0,07$	$22,15 \pm 0,24$	This study
250	$0,17 \pm 0,02$	$11,92 \pm 0,80$	[19]
258	$0,45 \pm 0,07$	$24,70 \pm 0,25$	This study
265	$0,55 \pm 0,02$	$34,19 \pm 0,80$	[19]

\*the  $n$  value after recalculation

In the temperature range of 230-243 °C a clear decrease of the  $n$  value is visible indicating the change of the diffusion type, which can be explained by the growing contribution of the grain boundary diffusion and the decrease of the phase boundary reaction contribution up to its leading to the Ag<sub>3</sub>Sn phase formation. At the temperature of 250 °C an anomalous decrease of the  $n$  value was noticed, which, however, cannot be clearly explained by the present research. At 265 °C the  $n$  value increases again and it may indicate the volume diffusion as the rate controlling factor.

Such a distinct the changes the exponential factor value  $n$  is involves in the changes of the rate constant  $k$ . When the volume diffusion predominates the obtained  $k$  values indicate an increase of the Ag<sub>3</sub>Sn phase layer growth rate. The examples of this can be seen at the temperatures of 258 °C and 265 °C (Table 2). Moreover, the  $k$  values are confirmed in by the morphology, because for the temperatures at which the  $n$  values where close to 0.5 a larger Ag<sub>3</sub>Sn phase width for long annealing periods of time was obtained.

TABLE 3

Values of the diffusion coefficients for Ag-Sn at different temperatures

<i>No.</i>	<i>Temperature</i> [°C]	<i>D</i> [m²/s]	<i>Diffusion coefficient</i>	<i>Literature</i>
1	20	1.00E-17	interdiffusion (diffusion couple)	[20]
2	120	7.34E-13	intrinsic, Sn in Ag <sub>3</sub> Sn	[21]
3	160	3.67E-12		
4		1.41E-14	interdiffusion	[22]
5	222	1.34E-14 <sup>+</sup>	intrinsic, Ag in Sn (tracer)	[23]
6	226	3.22E-12 <sup>++</sup>		
7	231	2.07E-09	intrinsic, Ag in Sn	[24]
8	243	8.37E-15*	interdiffusion, integrated	Own research
9		4.10E-15**		
10	258	8.85E-15*		
11		5.90E-15**		

<sup>+</sup>|| to the axis c in Sn tetragonal system

<sup>++</sup>⊥ to the axis c in Sn tetragonal system

\* Wagner model

\*\* numerical model [16]

Table 3 presents the values of the average interdiffusion coefficient obtained from the numerical model [16] and Wagner model for 243 °C and 258 °C as well as other values of the diffusion coefficients found in literature for the Ag-Sn system.

The obtained values of the average interdiffusion coefficient from Wagner model and the numerical model (Table 3) show certain difference in relation to each other. The difference of magnitude may result from taking into account in the numerical model calculations the points obtained from all the measurements instead of the average results for the given temperature including also those beyond the parabolic range. Moreover, the numerical model assumes that during the diffusion process the phase edges move, the concentrations are nonlinear and the component fluxes are discontinuous on the phases boundary.

The comparison of the obtained average interdiffusion coefficient from the numerical model and Wagner model (Table 3) with the values of the interdiffusion coefficients found in literature [20, 22] (Table 3, Nos. 1 and 4) indicates a good correlation with the value obtained at 160 °C. There are no available data in literature for the values of intrinsic coefficients of Ag and Sn in the Ag<sub>3</sub>Sn phase at the temperatures investigated in this study. The only data available in literature refer to lower temperatures for Sn in Ag<sub>3</sub>Sn [21] (Table 3, Nos. 2 and

3) and have a similar order of magnitude. The values from the numerical model and Wagner model (Table 3) can only be related to the intrinsic diffusion coefficients of Ag and Sn in the Ag<sub>3</sub>Sn phase.

Because of the lack of the data in literature concerning the intrinsic diffusion coefficients of Ag in Ag<sub>3</sub>Sn, the values of diffusion coefficients of Ag in Sn were also taken into account. The analysis of these values (Table 3, Nos. 5-7) in relation to Ag/Ag<sub>3</sub>Sn/Sn system (Table 3, Nos. 8-11) may indicate (at least in the initial stage of DS, before the Ag<sub>3</sub>Sn IP has been formed) a remarkable contribution of the diffusion of silver in comparison with the diffusion of tin in the Ag-Sn system. This is also confirmed by the results of Bruson et al. [24] who clearly indicate an increasing tendency of the diffusion rate of Ag in Sn together with the growing temperature.

The time necessary for DS in the case when the layer of solder is relatively thin (1-2 μm) due to magnetron sputtering or electrochemical deposition (Table 4), as is shown in the equation below (Eq. 2).

$$\tau = \frac{x^2}{2D} \quad (2)$$

in which x is the thickness layer Ag<sub>3</sub>Sn, D is the interdiffusion coefficient obtained from Wagner model or numerical model and τ is the time the growth layer Ag<sub>3</sub>Sn.

TABLE 4

Time ( $\tau$ ) needed to get assumption the  $\text{Ag}_2\text{In}$  thickness ( $x$ ) calculated from the coefficients ( $D$ ) obtained from Wagner model (W) and numerical model (N) by means of Equation 2

$x[\mu\text{m}]$	243 [°]		Model	258 [°]	
	$\tau$ [s]	$D$ [ $\text{m}^2/\text{s}$ ]		$D$ [ $\text{m}^2/\text{s}$ ]	$\tau$ [s]
1	60	8.37E-15	W	8.85E-15	57
2	240				227
1	122	4.10E-15	N	5.90E-15	85
2	488				339

Basing on the obtained results (Table 4) one can conclude that the DS process can be accomplished within reasonable time.

4. Conclusions

Diffusion soldering was successfully applied to form Ag/Sn/Ag joints at temperatures of 243 °C and 258 °C. The following main conclusions can be drawn based on the performed studies:

- 1° the only one intermetallic phase identified as  $\text{Ag}_3\text{Sn}$  exists in the joint in the form of scallops;
- 2° soldering time about 20 minutes is good enough to form  $\text{Ag}_3\text{Sn}/\text{Sn}$  and  $\text{Ag}_3\text{Sn}/\text{Ag}$  interfaces without voids, pores and cracks;
- 3° the formation of  $\text{Ag}_3\text{Sn}$  phase is governed by the volume diffusion at 258 °C ( $n = 0,45$ ) and by volume diffusion with some contribution of grain boundary diffusion at 243 °C ( $n = 0.38$ );
- 4° the calculated diffusion coefficients can be used in the production line to determine the time necessary to accomplish the soldering process.

REFERENCES

[1] W. C. Welch, J. Chae, K. Najafi, IEEE T. Adv. Packaging **28**, 643-649 (2005).

[2] P. Zięba, J. Wojewoda, Works of Technical Science Commission of the Polish Academy of Arts and Sciences **II**, 85-94 (In Polish) (2007).

[3] D. M. Jacobson, G. Humpston, Int. Mater. Rev. **51**, 313-328 (2006).

[4] S. Sommadossi, H. E. Troiani, A. F. Guillermet, J. Mater. Sci. **42**, 9707-9712 (2007).

[5] Program Pandat (database COST 531 v.2 (2006)).

[6] D. M. Jacobson, G. Humpston, Solder. Surf. Mt. Tech. **10**, 27-32 (1992).

[7] G. Humpston, D. M. Jacobson, S. P. S. Sangha, Diffusion soldering for electronics manufacturing. New Series 18, Endeavour, 55-60 (1994).

[8] <http://www.hkltechnology.com> (19.03.2009).

[9] X. Li, J. Cai, Y. Sohn, Q. Wang, W. Kim, Ag-Sn fluxless wafer bonding technology. International Conference on Electronic Materials and Packaging **11-14**, 1-6 (2006).

[10] <http://www.matcoinc.com> (19.03.2009).

[11] J. Wojewoda, P. Zięba, Mater. Eng. **138**, 11-23 (In Polish) (2004).

[12] H. K. Kim, H. K. Liou, K. N. Tu, Appl. Phys. Lett. **66**, 2337-2339 (1995).

[13] r. l. Su, L. C. Tsao, S. Y. Chang, T. H. Chuang, J. Mater. Eng. Perform. **11**, 365-368 (2002).

[14] T. L. Su, L. C. Tsao, S. Y. Chang, T. H. Chuang, J. Mater. Eng. Perform. **11**, 481-486 (2002).

[15] Y. W. Yen, W. T. Chou, Y. Tseng, C. Lee, C. L. Hsu, J. Electron. Mater. **37**, 73-83 (2007).

[16] J. Wojewoda, P. Zięba, Mater. Eng. **157-158**, 496-498 (In Polish) (2007).

[17] B. Pierragi, Oxid. Met. **27**, 177-185 (1987).

[18] F. J. J. Van Loo, rog. Solid State Ch. **30**, 47-99 (1990).

[19] Final report of the project No. 3 TO8C 028 29 financially supported by Ministry of Science and Higher Education, Institute of Metallurgy and Materials Science Kraków 2008 entitled "Structural and kinetic characteristics of electronic joints obtained in the diffusion soldering process" (In Polish).

[20] S. K. Sen, A. Ghorai, A. K. Bandyopadhyay, Thin solid films **155**, 243-253 (1987).

[21] C. M. Chen, S. W. Chen, Acta mater. **50**, 2461-2469 (2000).

[22] H. F. Zou, Z. F. Zhang, J. Alloy. Compd. **469**, 207-214 (2009).

[23] B. F. Dyson, J. Appl. Phys. **37**, 2375-2377 (1966).

[24] A. Bruson, M. Gerl, Phys. Rev. **B 21**, 5447-5454 (1980).