Study of Undercooling and Recalescence During Solidification of $Sn_{62.5}Pb_{36.5}Ag_1$ and $Sn_{96.5}Ag_3Cu_{0.5}$ Solders in Real Electronic Joints

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Undercooling and recalescence were studied using the differential scanning calorimetry (DSC) method on real electronic systems. Two solder pastes, $Sn_{62.5}Pb_{36.5}Ag_1$ and $Sn_{96.5}Ag_3Cu_{0.5}$, were used for preparation of electronic joints. Various combinations of these solders and soldering pads with different surface finishes such as Cu, Cu-Ni-Au, Cu-Sn, and $Cu-Sn_{99}Cu_1$ were used. During melting of both pastes, the Sn and $Sn_{99}Cu_1$ surface finishes immediately dissolved in the solder and the Cu surface coating was exposed to the melt. Therefore, practically the same undercooling was found for the Cu, Cu-Sn, and $Cu-Sn_{99}Cu_1$ coatings. The lowest undercooling was found for the Cu-Ni-Au surface finish for both solder pastes. If two separated electronic joints were made on the sample, two separate peaks were found in the DSC signal during solidification. In the sample with only one joint, only one exothermic peak was found. These findings were observed for all paste/surface finish combinations. These data were analyzed, showing that this effect is a consequence of undercooling and recalescence: Latent heat released during solidification of the joint increases the surrounding temperature and influences all the processes taking place.

Key words: Undercooling, Sn-based solders, recalescence, DSC

INTRODUCTION

Sn-Pb solder has long been used in the electronics industry, but because of its toxic nature, certain restrictions have been imposed on its use by the European Restriction of Hazardous Substances (RoHS) directive, and therefore many researchers have looked at replacing it. Pb-free solders are mostly Sn-containing binary and ternary alloys.[1] A special class is formed by eutectic alloys of Sn with noble metals such as Ag and Cu. Eutectic alloys are used as solders because they have a single, low melting point. Most Pb-free alloys have a higher melting point compared with eutectic Sn-Pb. The reflow temperature is higher by about 30°C. This increases the dissolution rate and solubility of Cu and Ni. During reflow, the eutectic solder liquid comes into contact with electronic parts (Cu, Ni, Au, Sn). Intermetallic compounds grow in the interface and drain Sn from the solder liquidus. As an example, we mention the intermetallic compound $AuSn_4$, formed from four atoms of Sn and one of Au. The interfacial reactions in electronic joints have been studied, for example, by Ho et al.[2]

A Sn-based joint, together with the printed circuit board (PCB) and electronic components, creates a very complex system where various materials with different physical properties come into contact. The PCB, soldering pads, eutectic Sn-based alloy, and parts of the electronic components interact at temperatures higher than the melting point of the solder. Chemical reactions occur in the liquid state not only at the various interfaces but even in the melt,

depending on the solubility of single surface components in the liquid Sn-based alloy (flux reactions). Components of the PCB face finish and of the metal parts of electronic components create intermetallic compounds with Sn. This process deprives the liquid solution of Sn, and the original eutectic alloy becomes noneutectic. This means that the solid solution in the joint is not fully homogeneous. It is evident that each component of this complex and heterogeneous system has different thermal conductivity and thermal expansion, which determine the final physical properties of the joint. Different temperature gradients lead to inhomogeneous thermal fields on the PCB, and different thermal expansion coefficients in adjacent components lead to the development of internal stresses.

Similar to many metals and alloys, Sn and its alloys are prone to undercooling prior to initial nucleation of solid from the melt. This means that a solid phase does not begin to form during cooling at the temperature given by the phase diagram. Nucleation occurs on foreign crystals, for example, ceramic grains of the crucible, oxide on the melt surface, or fine particles dispersed throughout the liquid. Nucleation temperatures can vary from a fraction of a degree to several hundred degrees below liquidus, depending on the alloy system and other factors. It is therefore evident that electronic joints where intermetallic compounds exist in the interface between the electronic parts and solder have very different nucleation centers compared with the pure alloy when studied alone. In our previous study,[3] we examined pure alloys $Sn_{96}Ag_4$, $Sn_{99}Cu_1$, $Sn_{97}Cu_3$, $Sn_{96.5}Ag_3Cu_{0.5}$, $Sn_{95.5}Ag_{3.8}Cu_{0.7}$, and $Sn_{63}Pb_{37}$ using the differential scanning calorimetry (DSC) method. The hysteresis was smallest for the $Sn_{96.5}Ag_3Cu_{0.5}$ alloy (9.6°C) and greatest for $Sn_{63}Pb_{37}$ (28°C). This significant difference is evidently a consequence of the fact that, in the Sn-Pb system, intermetallic compounds do not create nucleation centers for solidification. It is assumed that these values for the difference between the melting point and solidification temperature will not be valid in real electronic joints where intermetallic compounds will occur in the interfaces. In our study, it was also shown that the solidification temperatures for all the studied alloys depended on their thermal history, i.e., the maximum thermal cycle temperature and heating/cooling rates. Solidification took place practically immediately in a large part of the sample. The rate of this process did not depend on the composition or thermal history of the material.

The undercooling effect in Sn alloys leads to instant release of latent heat. The temperature of the sample and therefore also of its surroundings suddenly increases (recalescence). The temperature increase of pure Sn, as measured by a thermocouple immersed in 163.2 mg of melt,[4] was about 30°C. In the case of a PCB, the various temperature gradients can be expected to depend on the thermal

conductivity of the surroundings. The electronic components are connected by Cu, which is the second best thermal conductor among the metals. It is necessary to bear in mind that the PCB system with solder joints and electronic components is a heterogeneous system in which, during solidification, local heating can occur at places where several electronic parts are located. It was shown that the solidification temperature depends on the thermal history of the melt. The thermal history of individual joints may not be the same, and we can expect various solidification conditions for individual joints.

The high solidification rate after nucleation may be a cause of heterogeneity in the joint.[5] The heterogeneity of the joint is then connected with the internal stresses because of the difference in the thermal expansion of individual components and the different temperature gradients in the joint during solidification. A consequence of these effects may be lower joint lifetime, as well as undesired effects such as the tombstone effect,[6,7] void formation,[8,9] nonspreading effect,[10,11] flux spattering,[12,13] dewetting,[14] whisker growth,[15,16] etc.

The complexity of PCBs has dramatically increased over the last three decades. PCBs are highly complex and multilayered with a mixture of through-holes and soldering pads. Board layouts have consequently increased in density with tighter tolerances and decreased distances between electrical contacts. Therefore, study of undercooling and recalescence is crucial. It has been well recognized that material properties obtained from bulk solder materials are no longer applicable for estimation of the reliability of microscale solder interconnects. This study aims to understand the solidification process of $Sn_{62.5}Pb_{36.5}Ag_1$ and $Sn_{96.5}Ag_3Cu_{0.5}$ solders in an electronic component–solder–PCB system, in order cognition, and the influences on this process. This study also aims to examine the recalescence effect and its influence on solidification processes in real electronic systems.

EXPERIMENTAL PROCEDURES

In the present study, commercially prepared solder pastes with composition (wt.%) $Sn_{62.5}Pb_{36.5}Ag_1$ (P1; Cobar Co., Breda, The Netherlands) and $Sn_{96.5}Ag_3Cu_{0.5}$ (P2; Kester Co., Itasca, IL) were used for preparation of real joints. Samples were prepared using PCB substrate with thickness of 0.5 mm. The samples were circular with diameter of 3.2 mm so that it was possible to place them at the bottom of the crucible in the DSC device. Pastes were used to connect the PCB substrate to a real electronic component. PCB substrates that are in contact with solder are known as soldering pads. The base of the soldering pad is nearly always Cu. Samples for DSC measurements were prepared from commercially prepared PCB with a Cu layer of thickness 50 μm. As a further surface finish, the Cu soldering pad was

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coated with electroless Ni of 5 μm thickness followed by immersion Au of 0.1 μm thickness (Cu-Ni-Au), with immersion Sn of 1 μm thickness (Cu-Sn), or with hot-air solder leveling to deposit $Sn_{99}Cu_1$ alloy with 6 μm to 8 μm thickness (Cu-$Sn_{99}Cu_1$). These samples were also obtained commercially.

The first series of DSC measurements were carried out on circular-shaped samples of fully laminated PCB substrate with Cu, Cu-Ni-Au, Cu-Sn or Cu-$Sn_{99}Cu_1$ alloy surface finish. At the center of the circular sample, paste was printed with dimensions of 2 mm × 1 mm through a stencil (stencil thickness 100 μm). This sample was placed into the DSC crucible. These samples are denoted as PCB-O/Cu, PCB-O/Cu-Ni-Au, PCB-O/Cu-Sn, and PCB-O/Cu-$Sn_{99}Cu_1$. The second series of samples for DSC measurements were prepared from circular-shaped samples of PCB with two soldering pads with dimensions of 0.5 mm × 0.5 mm at a separation of 1 mm, defined for a 0603 surface-mounted device (SMD) resistor. Figure 1 shows this sample. The surface finishes of these pads were the same as in the previous case. A 0603 SMD-type resistor was soldered to the soldering pads on the PCB samples using paste P1 or P2. These samples are denoted as R-PCB/Cu, R-PCB/Cu-Ni-Au, R-PCB/Cu-Sn, and R-PCB/Cu-$Sn_{99}Cu_1$. The third series of samples were obtained by connecting two PCB circular samples with the same surface finish as in the previous cases. The samples were connected only using paste P1 at the soldering pad locations. These samples are denoted as PCB-T/Cu, PCB-T/Cu-Ni-Au, PCB-T/Cu-Sn, and PCB-T/Cu-$Sn_{99}Cu_1$.

DSC measurements were carried out on paste P1 and P2 and on all combinations of PCB samples described above. Measurements were performed under He protective atmosphere using a SETSYS Evolution 24 instrument (Setaram Co., Caluire, France) at heating/cooling rates of 10 K/min for two or three thermal cycles. The maximum thermal cycle temperature was 220°C for paste P1 and 250°C for paste P2.

RESULTS AND DISCUSSION

Solders $Sn_{62.5}Pb_{36.5}Ag_1$ and $Sn_{96.5}Ag_3Cu_{0.5}$ in paste form were studied by DSC. Figure 2 shows the results. The second runs are presented because the temperature dependence of the heat flow in the first runs was influenced by evaporation of the binder. $Sn_{62.5}Pb_{36.5}Ag_1$ showed lower undercooling (9°C) than $Sn_{96.5}Ag_3Cu_{0.5}$ (15°C). The undercooling is defined as the temperature gap between the melting onset temperature and the solidification onset temperature. A weak size effect was found for both solders: the undercooling increased as the size of the solder decreased. This effect was ~1°C for both solders (6.5 mg was compared with 65 mg of solder paste). Such a size effect was also demonstrated, for example, in Refs. 17–19. Zhou et al.[19] studied Sn-3.0Ag-0.5Cu alloy solder balls and Sn-3.0Ag-0.5Cu/Cu joints by DSC as a function of solder size. Smaller samples showed greater undercooling and only a single solidification peak. Larger samples exhibited double solidification peaks and less undercooling. The double peaks were interpreted based on the easier nucleation of β-Sn phase in comparison with the more difficult nucleation of the eutectic phase.

The DSC data for pure solders P1 and P2 were compared with the results obtained for these solders applied on PCBs with various surface finishes (PCB-O-type samples). Figures 3 and 4 reveal lower undercooling for the PCB-O-type samples than for the pure solders. Also, the amount of solder paste used was smaller for the PCB-O samples: 2 mg instead of 6.5 mg. No size effect was found in these experiments because the influence of the interface was dominant. The interface with intermetallic compounds creates nucleation centers, which increase the
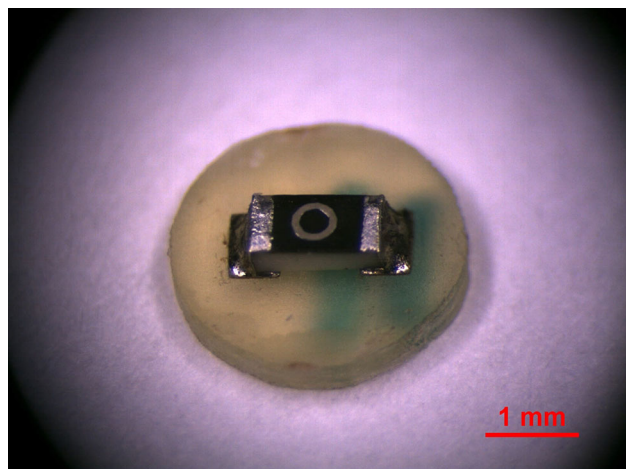

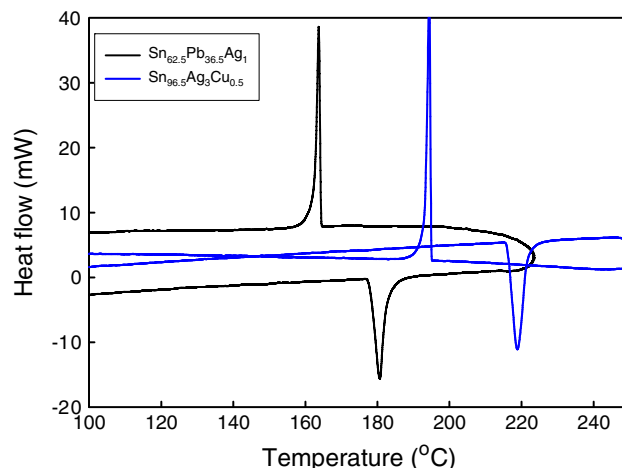Fig. 1. Sample with 0603 SMD resistor soldered to the soldering pads on the PCB.

Fig. 2. Temperature dependence of heat flow for $Sn_{62.5}Pb_{36.5}Ag_1$ and $Sn_{96.5}Ag_3Cu_{0.5}$ solders.
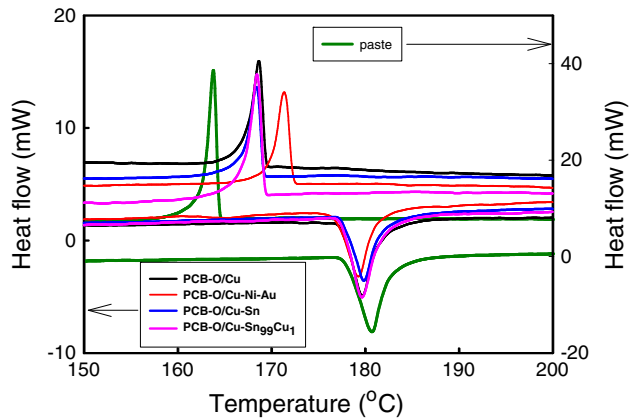
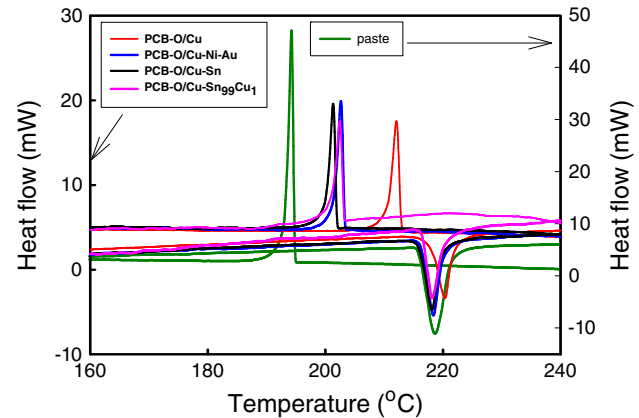Fig. 3. Temperature dependence of heat flow for solder P1 for samples PCB-O/Cu, PCB-O/Cu-Ni-Au, PCB-O/Cu-Sn, and PCB-O/Cu-Sn$_{99}$Cu$_1$.

Fig. 4. Temperature dependence of heat flow for solder P2 for samples PCB-O/Cu, PCB-O/Cu-Ni-Au, PCB-O/Cu-Sn, and PCB-O/Cu-Sn$_{99}$Cu$_1$.

solidification temperature and therefore decrease the undercooling.

Figures 3 and 4 also show the influence of the different surface finishes on the solidification temperature (undercooling). All the surface finishes used in this study had the same base, namely soldering pad with Cu coating. When soldering pads with Sn or Sn$_{99}$Cu$_1$ surface finish were exposed to the melt, the Sn or Sn$_{99}$Cu$_1$ alloy immediately dissolved and the Cu coating was exposed to the melt. Figure 5 shows the temperature dependence of the heat flow when a mixture of low-temperature paste P1 and high-temperature paste P2 was heated. It can be seen that the high-temperature solder P2 dissolved into the liquid solder P1 in the temperature range of melting of this alloy; the original eutectic alloys P1 and P2 create a new, noneutectic alloy, as revealed by the two peaks seen in the solidification process in Fig. 5. Similar processes may be expected in real joint systems when the surface finish is made of Sn or Sn-based solder. If the originally eutectic solder becomes noneutectic, there will be an increase of joint inhomogeneity.

The interfacial reactions between the molten solder and the Cu, Cu-Sn, and Cu-Sn$_{99}$Cu$_1$ surface finishes take place by the same mechanism because Cu is exposed to the melt in each case. We can see that the undercooling has the same character for all three surface finishes and for both soldering pastes. Cu-Sn is an important system with a 1000-year history; therefore, much information can be found in literature. From many literature sources it is obvious that Cu$_6$Sn$_5$ intermetallic compound occurs in the interface between liquid Sn-based solder and Cu.

The case of the fourth surface finish, Cu-Ni-Au, is more complicated. When the solder is melting, Au rapidly dissolves in the bulk solder, and precipitates of AuSn$_4$ are found in the solder after cooling. Ni is exposed to the solder, and the binary Ni$_3$Sn$_4$ compound occurs in the interface. This helps Cu diffusion and the formation of (Au,Ni)$_6$Sn$_4$ intermetallic
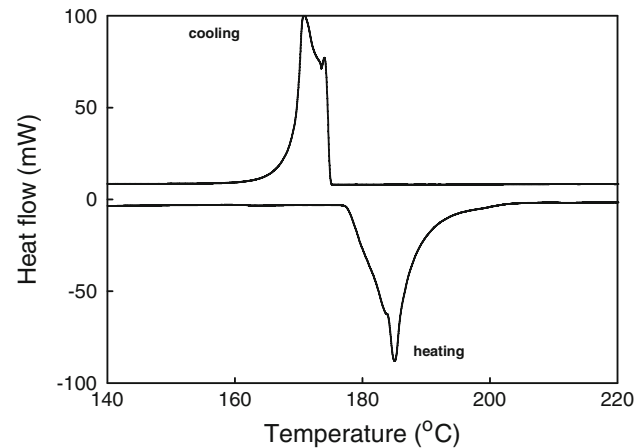

Fig. 5. Temperature dependence of heat flow for a mixture of solders P1 and P2 (mass of P1 paste 67.2 mg, mass of P2 paste 16.9 mg).

compound at the interface.[20] The intermetallics at the interface depend on further factors such as the kind of solder and the thermal history of the system, for example, the number of reflows.[21] Our results show that this type of interface leads to lower undercooling than an interface with intermetallic Cu$_6$Sn$_5$ compound. This conclusion is valid for both studied solders, even though the solubility of Cu in Sn-Pb-based solders is lower than in Sn-Ag-Cu-based solders[22] and redistribution of (Au,Ni)$_6$Sn$_4$ on Ni$_3$Sn$_4$ does not occur in Pb-free solders by the same mechanism as in Pb-Sn-based solders.[22]

Application of electronic components to a PCB requires a minimum of two solder joints. Figure 1 shows the sample with the electronic components. Figures 6 and 7 show the temperature dependences of the heat flow for the R-PCB/Cu samples with solder pastes P1 and P2. Table I presents the solidification temperatures for the R-PCB samples with all the used surface finishes and for solder paste P1 and P2. Two runs are presented in the
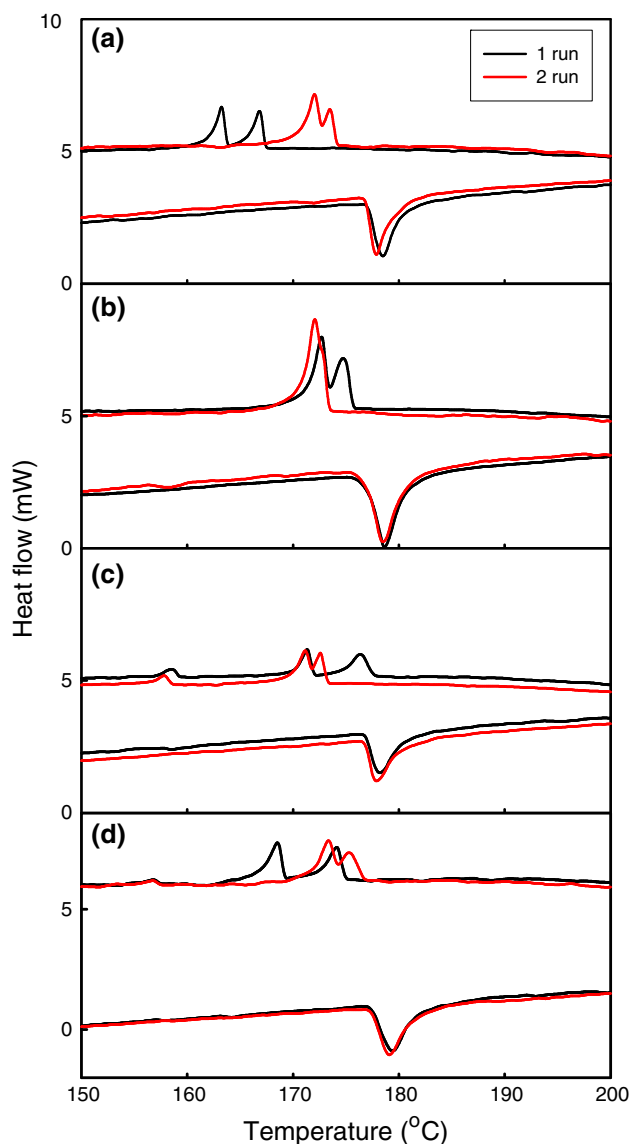
Fig. 6. Temperature dependence of heat flow for solder P1 for sample R-PCB/Cu.
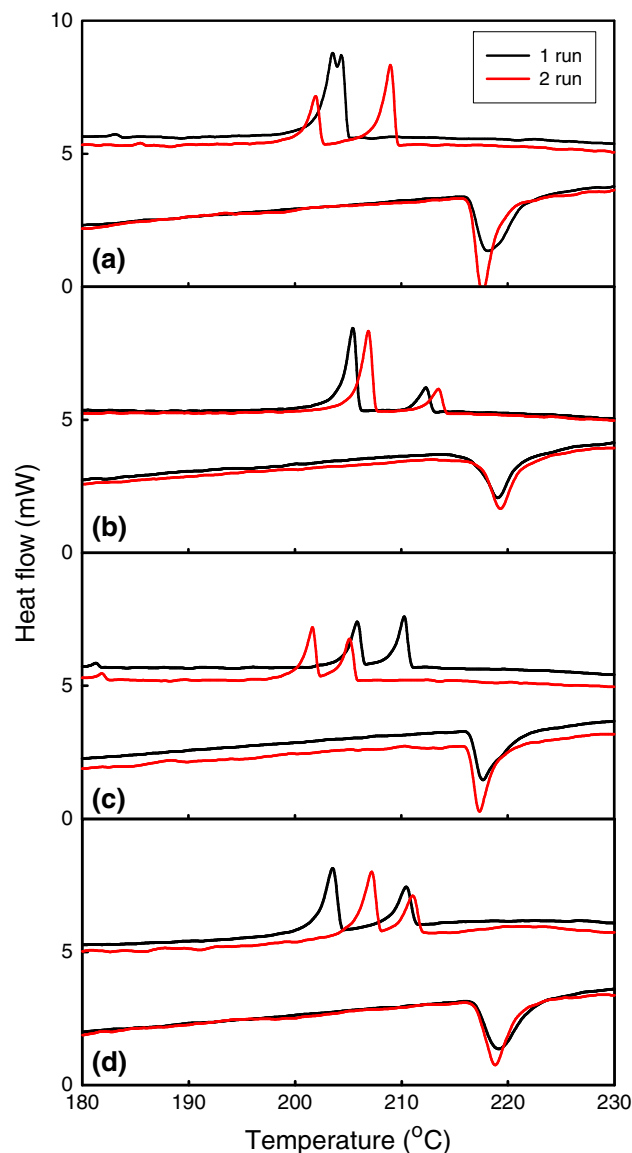

Fig. 7. Temperature dependence of heat flow for solder P2 for sample R-PCB/Cu.

figures as well as in the tables. Two peaks or double peaks were found in all cases. This finding is surprising, because only one peak was found during melting in all cases. However, double peaks are often presented in the literature[23,24] and are assigned to the noneutectic behavior of the studied Sn-based systems. Figures 3 and 4 show DSC results where the solder behaves as a eutectic alloy even when connected to a PCB with various surface finishes (Sn can be consumed or enters the liquid from the surface finish). No double peak in the heat flow during solidification was found for this type of measurement. The changes of composition are too small to lead to noticeable double solidification peaks. The two peaks and double peaks presented in Figs. 6 and 7 and Table I cannot be assigned to noneutectic behavior of the solders. All these results show that the solidification of the two solder joints

does not take place at the same time. It has been reported[5,9] that undercooling leads to a very high rate of the solidification process that is connected with a sudden release of latent heat. The released heat warms the surroundings of the joint and evidently increases the temperature of the second joint. Therefore, solidification of this joint takes place later. Solidification of solder is connected with volume contraction. While sudden solidification of one joint of an electronic component takes place, the second joint remains in the liquid state. This can lead to a deviation of the electronic component from its original position and to the creation of voids or various effects, for example, the tombstone effect. The presented results also show that the hysteresis in the DSC results is lower for solders connected to electronic components than for pure solders without the electronic component. The temperature difference

Table I. Solidification temperature T_1 (onset of the first peak in the temperature dependence of the heat flow) and T_2 (onset of the second peak in the temperature dependence of the heat flow) for $Sn_{62.5}Pb_{36.5}Ag_1$ and $Sn_{96.5}Ag_3Cu_{0.5}$ solders and R-PCB samples with different surface finishes

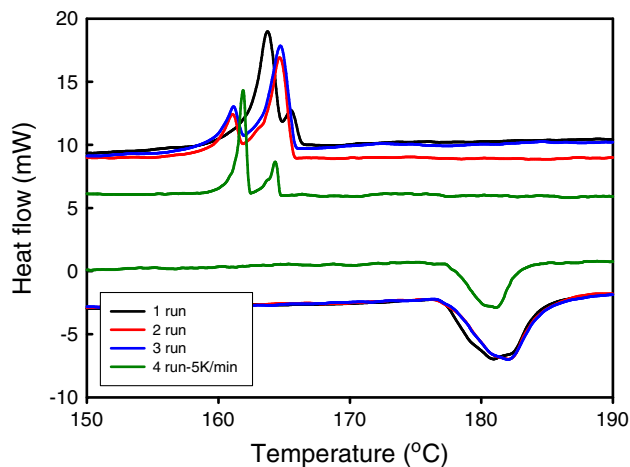| Surface Finish | No. of Runs | $Sn_{62.5}Pb_{36.5}Ag_1$ | | $Sn_{96.5}Ag_3Cu_{0.5}$ | |
		T_1 (°C)	T_2 (°C)	T_1 (°C)	T_2 (°C)
Cu	1	167.4	163.9	205.1	204.0
	2	174.2	172.8	209.7	202.7
Cu-Ni-Au	1	175.9	173.5	213.2	206.2
	2	173.6	172.6	214.3	207.8
Cu-Sn	1	177.7	172.1	211.0	206.6
	2	173.4	171.8	205.9	202.4
Cu-Sn$_{99}$Cu$_1$	1	175.0	169.2	211.3	204.3
	2	176.9	174.3	211.9	208.0

Fig. 8. Temperature dependence of heat flow for two spots of solder P1 on a PCB without surface finish (in the fourth thermal cycle, the sample was rounded at about 180°).
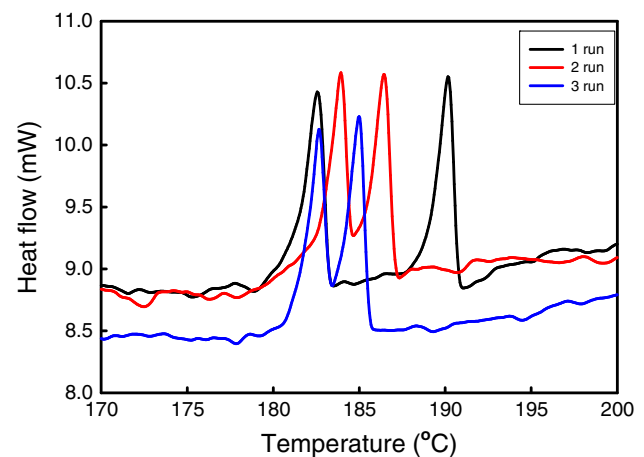

Fig. 9. Temperature dependence of heat flow (cooling) for two spots of solder P2 on a PCB without surface finish.

between the two solidification peaks is quite random. This difference depends on the thermophysical and shape properties of the whole system.

To elucidate the existence of the two peaks, two spots of paste P1 of several milligrams were applied to a PCB disk without surface finish. In this experiment, the amount of paste was not the same for the two spots. The sample was placed in the crucible such that the line connecting the two spots was perpendicular to the line connecting the reference and measuring crucibles. The results obtained in four thermal cycles are shown in Fig. 8. Two peaks were found in all thermal cycles. We assume that the smaller peak corresponds to release of latent heat by the smaller solder spot and the larger peak corresponds to release of latent heat by the larger solder spot. The order of the peaks was opposite in the first thermal cycle compared with the following two thermal cycles. The opposite order of peaks was also obtained when a cooling rate of 5 K/min was used. It is assumed that the change of the peak order is a consequence of a change in the

heat transfer on the sample in individual thermal cycles. The heat flow in the first thermal cycle can be influenced by the binder from the paste. A heating/cooling rate of 5 K/min leads to a different temperature distribution on the sample compared with the case of 10 K/min. A similar experiment was carried out with paste P2 with two spots on a PCB disk printed through a stencil (two spots of dimensions 0.5 mm × 0.5 mm with separation of 1 mm). The DSC results (Fig. 9) once more show two peaks with the same area (the solder spots were practically the same). These experimental results clearly confirm the hypothesis that, if two peaks are found in the temperature dependence of the heat flow for two separated solder joints, then these joints solidify at different times. This does not mean that they solidify at the same temperature, because undercooling is very sensitive to many factors. This conclusion applies even for the more complex system of the R-PCB samples. It is generally known that the temperature gradient in a cylindrical furnace during linear heating/cooling decreases from the furnace walls towards the furnace center. The
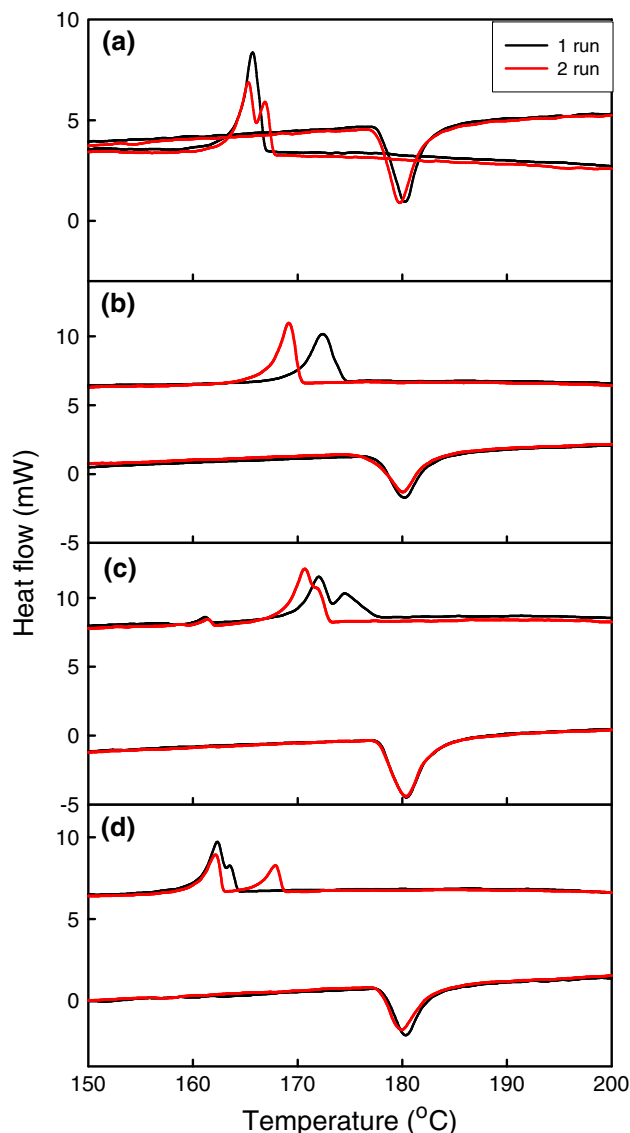
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Fig. 10. Temperature dependence of heat flow for solder P1 in sample PCB-T/Cu.

Table II. Solidification temperatures T_1 (onset of the first peak in the temperature dependence of the heat flow) and T_2 (onset of the second peak in the temperature dependence of the heat flow) for $Sn_{62.5}Pb_{36.5}Ag_1$ solder and PCB-T samples with different surface finishes

Surface Finish	No. of Runs	$Sn_{62.5}Pb_{36.5}Ag_1$	
		T_1 (°C)	T_2 (°C)
Cu	1	167.0	–
	2	167.8	166.0
Cu-Ni-Au	1	174.6	–
	2	170.5	–
Cu-Sn	1	177.6	173.0
	2	173.0	171.5
Cu-Sn$_{99}$Cu$_1$	1	164.2	163.1
	2	168.7	162.8

intermetallic compounds with Cu and Sn. This increases the solidification temperatures, as seen in Figs. 3 and 4.

The release of latent heat leads to local preheating, an effect that could influence the reliability of electronic packaging on PCBs. Lee[25] showed that processes taking place during the cooling stage of the reflow process are the main factors affecting cracking defects. A slow cooling rate leads to weak fatigue resistance, whereas a fast cooling rate can cause internal cracking of solder joints and delamination of bond pads. Modeling and optimization of the reflow thermal profile process was presented by Tsai,[26] who demonstrated that not only the temperature setting and flow speed but also the board configuration are important for the solidification process. The effect of the solder joint arrangement on the solidification of individual joints was shown in the study of Lau et al.,[27] which focused on the development of a numerical approach to investigate the temperature and thermal stress in various joint arrangements during joint solidification. The temperature, as well as the thermal stress, is influenced by the release of latent heat. The importance of the problem of the solder joint arrangement on the PCB and the release of latent heat increases with their miniaturization. The results of this work show that study of thermal analysis can be an important method for investigating this very complicated problem where unsteady thermal conduction, as well as mass diffusion processes, takes place. This study opens many further questions and problems that can be solved by this method.

temperature distribution in the sample will be dependent on this temperature gradient, on the position of the sample in the furnace, on the thermophysical properties of the sample, and on the interior arrangement of the apparatus.

Figure 10 shows the DSC data obtained for the third sample type: PCB-T with solder P1 and Cu surface finish. Table II presents the solidification temperatures for two runs. In this series, we found three cases where only one peak occurred in the temperature dependence of the heat flow during solidification. We assume that solidification of both joints took place at the same time. Comparison of the heat flow measurements for the R-PCB and PCB-T samples leads to the conclusion that part of the electronic component in the solder joint decreases the undercooling. The soldered part of the electronic components contains Ni, which can form

CONCLUSIONS

1. $Sn_{62.5}Pb_{36.5}Ag_1$ and $Sn_{96.5}Ag_3Cu_{0.5}$ solder pastes show undercooling, being higher for the Pb-free solder $Sn_{96.5}Ag_3Cu_{0.5}$.
2. The influence of the surface finish of the soldering pads on the solidification process depends on the

kind and amounts of intermetallic compounds that initiate nucleation as the first step of the solidification process. The Sn and $Sn_{99}Cu_1$ surface finishes do not influence the solidification process, because they immediately dissolve in the molten solder, leading to interaction of the underlying Cu with the melt. Our results show that Cu-Sn intermetallic compounds lead to greater undercooling than binary Sn-Ni or ternary Sn-Ni-Cu intermetallic compounds that occur when Au-Ni coating enters the molten solder.

3. This study using the DSC method applied to real electronic joints has led to new knowledge regarding the behavior of solder joints in electronic assembly systems. Our results show that the undercooling process is very sensitive not only to the material properties of the system but also to its thermal properties. Two solidification peaks were found during cooling of real electronic components soldered to a PCB at two separate locations. The time shift between the solidification of the two separated joints is a consequence of recalescence. The influence of the chemical composition of the two solder pastes on the two peaks was negligible. Our results show the necessity of studying thermal transport for real PCBs with soldering pads, electronic components, and connections between them.

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