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Joining of lead–antimony and lead–calcium alloy lugs by a laboratory cast-on-strap process

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Abstract

A laboratory-scale characterization of the cast-on-strap joining process has been conducted using an existing lead–antimony strap alloy and low-antimony lead and lead–calcium alloy lugs. Increasing mould temperature in the range from 150 to 250 °C only marginally decreases the initially rapid cooling rate of the strap melt, but considerably decreases the subsequent cooling rate when the strap melt starts to solidify. Increasing the melt pouring temperature from 500 to 550 °C has little influence on the overall cooling characteristics of the strap. Joining of the lug/strap couple can be characterized by lack of fusion, partial fusion and complete fusion from the strap surface to the lug tip. At a mould temperature of 150 °C, a significant lack of fusion and considerable porosity are observed. The degree of fusion increases significantly when the mould temperature is increased from 150 to 200 °C. Pretinning effectively eliminates lack of fusion and considerably reduces the level of porosity. This most likely results from the presence of lower melting point material between the lugs and strap. There is good fusion and low porosity for lead–calcium alloy joints for which a more aggressive acid-based flux is used.

Introduction

In a lead/acid battery, cast-on-strap (COS) joints are currently used to group the individual plates in each cell. These joints support the cells in a battery and hence can be under a high shear stress. These highly stressed joints and the highly aggressive environment inside a battery can result in the easy growth of cracks in the joints if there are defects, such as lack of fusion, that can serve as initial cracks. Lack of fusion and a high level of porosity in the joints can prevent good electrical contact between the plates and the terminals that, in turn, will limit the performance of a battery. Extensive cracking of the joints can cause a complete battery failure and hence the quality of COS joints is critically important for the performance of a battery.

As noted by Prengaman [1], the Battery Council International Technical Committee performed a failure analysis of automotive batteries that showed the major causes of manufacturing defects to be the joining of the lugs to the straps and the through-the-partition welds. Hence, aspects of COS joints have been discussed extensively in

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the past few years [1–5]. These investigations indicate that the COS process is poorly understood with respect to a detailed description and understanding of joint quality. Accordingly, Pasminco Research Centre has been conducting a research programme with the following objectives:

(i) to gain an improved understanding of the COS process in order to provide better technical service to battery manufacturers so that premature failure of automotive batteries is minimized;

(ii) to use this improved understanding to develop improved COS alloys.

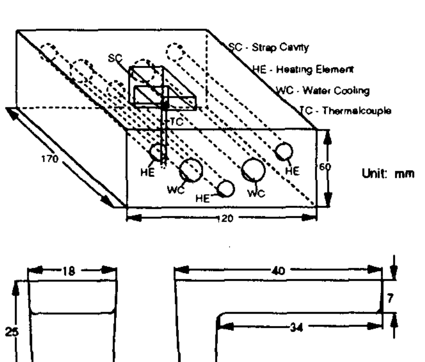
This paper describes the first phase of the programme on the development of an improved understanding of the COS process. This involves the simulation of the COS process in a laboratory-scale operation in which the thermal history of the strap melt is measured and COS joints are made. Joint quality is then examined with a metallographic method and is defined in terms of the degree of fusion and the extent of porosity.

Experimental

Cast-on-strap facility

Figure 1(a) shows the design of the strap-moulding system. The mould was made of low carbon steel with heating elements placed on each side of, and underneath, the strap cavity. A thermocouple was placed close to the bottom of the strap cavity to measure the mould temperature. The latter was controlled through an auto-setting proportional integral differential (PID) in a microprocessor-based temperature controller. The detailed dimensions of the cavity are shown in Fig. 1(b). The vertical portion of the strap simulates the part used for the through-the-partition joining.

A jig was designed to hold five lugs in a group with an interlug spacing of about 4 mm. A mechanism was attached to allow the group of lugs to be dipped into the strap melt, as illustrated schematically in Fig. 2. Dipping was performed manually. Figure 3 shows the strap-moulding system. To produce the lugs, another mould was made. This was based on the similar design to that reported in previous work [6, 7]. The lugs had the dimensions: 40×12×1.5 mm. The facility for melting the strap alloy was as described previously [6, 7].



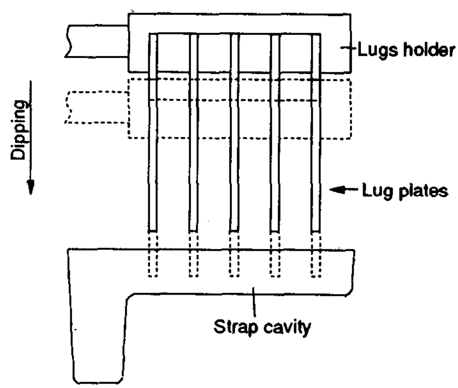


Fig. 1. Design details of: (a) strap mould; (b) dimensions of strap cavity.

Fig. 2. Schematic of lug holder and dipping action simulation of the COS process.

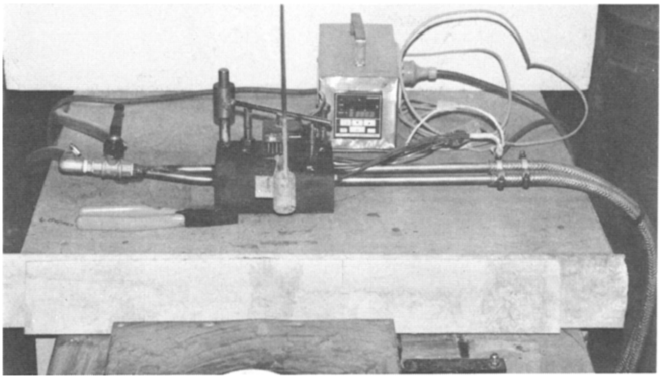


Fig. 3. Apparatus for making joints in the COS joining process.

TABLE 1

Composition (wt.%) of lug alloys and strap alloy and tinning metal (the balance is lead)

Alloy	Sb	As	Cu	Se	S	Ca	Sn	Al	Bi
S20	1.7	0.23	0.052	0.019	0.0016				0.013
D04						0.10	0.29	0.014	0.022
S31	2.7	0.045	0.011	<0.005	<0.005		0.03		0.019

Alloys and materials

A low-antimony–lead alloy (S20) and a lead–calcium alloy (D04) were used to cast the lugs. Their compositions, as analyzed in previous studies [6, 7], are listed in Table 1. The strap alloy (S31) was analysed by the direct-current plasma method; the composition is also given in Table 1. Commercially pure tin was used for lug pre-tinning.

Two kinds of fluxes were employed. These were made and supplied specifically for the COS process by TBS Engineering Ltd., UK. As recommended by TBS Engineering [8], TBS no. 5 flux (oil-based) was used for the low-antimony lugs and TBS no. 6 flux (acid-based) for the lead–calcium counterparts. A very thin coating of sodium silicate liquid was applied to the cavity surface after every three castings. Any excess sodium silicate was carefully removed with a cotton bud.

Temperature measurement

Before making any COS joints by dipping the lugs into the molten strap metal, the thermal history of the strap metal, after pouring into the strap cavity, was determined by simple temperature measurements. The tip of a K-type thermocouple (1 mm diameter) was placed at the centre of the horizontal section of the strap cavity. The output of the thermocouple was transferred to an Osborne SX-16 computer with a PCL-818-CS data acquisition system which recorded four readings every second. Data were obtained using melt pouring temperatures of 500 and 550 °C and mould temperatures of 150, 200 and 250 °C.

Initially, S20 alloy was used to develop the procedure for pouring molten strap alloy into the strap cavity. The use of a manual procedure meant, however, that control of the depth of the strap melt was inconsistent. During the later stages of the

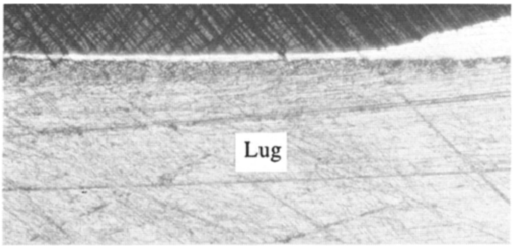


Fig. 4. Micrograph showing the thin tin coating on a low-antimony alloy lug. (Note: tin coating is significantly thicker at tip of the lug, 100×.)

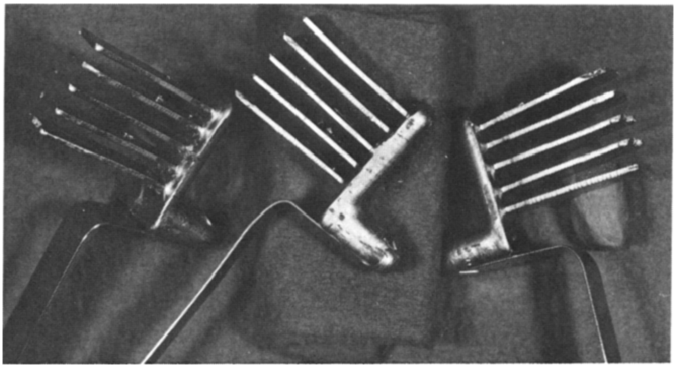


Fig. 5. Samples of COS joints.

development of the pouring procedure, thermal measurements were also made for this S20 alloy to provide preliminary data.

Joining procedure

Lugs were cast with a melt pouring temperature of 470 °C and a mould temperature of 240 °C and the samples were air-cooled after casting. Flux was applied by lightly wiping the lug surface using a cotton bud soaked with flux. For lug pretinning, a thorough investigation to establish the best tinning procedure was not made. Prefluxing and dipping in a tin melt for 5 s at 250 °C was found, however, to be satisfactory for the production of a tin coating on the lead alloy lug, as shown in Fig. 4.

To make COS joints, five lugs were grouped and positioned, as illustrated in Fig. 2 (solid lines). The bottom ends of the lugs were about 5 mm above the surface of the mould cavity. A specially-made casting cup (Fig. 3) was used to transfer the strap melt at 500 °C from the furnace and to pour this melt into the strap cavity. During pouring, the tip of the casting cup was placed very close to the strap cavity. The pouring operation took ~3 to 4 s. Immediately after pouring, the group of lugs was pushed down into the melt. The time interval between the finish of pouring to the finish of dipping was considerably less than 1 s. In a COS machine, the time interval between the melt being poured into the strap cavity and dipping of the lugs is normally set at ~ 0.5 s [8].

After the strap alloy had completely solidified, the mould was allowed to cool by turning off the heating elements and turning on the mould water-cooling (Fig. 1(a)). After taking out the joint, the mould was reheated to the desired temperature for the production of the next joint. Three COS joints each were made for: (i) low-antimony–lead alloy lugs without pretinning; (ii) low-antimony–lead alloys lugs with pretinning; (iii) lead–calcium alloy lugs without pretinning. Samples of COS joints are shown in Fig. 5.

Assessment of joint quality

An individual lug dipped into a strap is illustrated schematically in Fig. 6. Figure 6(a) represents the 'imaginary' extreme case of complete lack of fusion (solid line between the lug and the strap). This can result from the complete lack of wetting between the couple and no lug melting. In a real case, as shown later, the lug/strap joint couple will have different degrees of fusion from the strap surface to the lug tip, as illustrated in Fig. 6(b). In this Fig., the solid line still represents a lack of fusion whilst the broken line identifies the zone of partial fusion and complete fusion. The length of the lug/strap interface over which lack of fusion occurs has been measured as an important criterion for quality assessment of the joint.

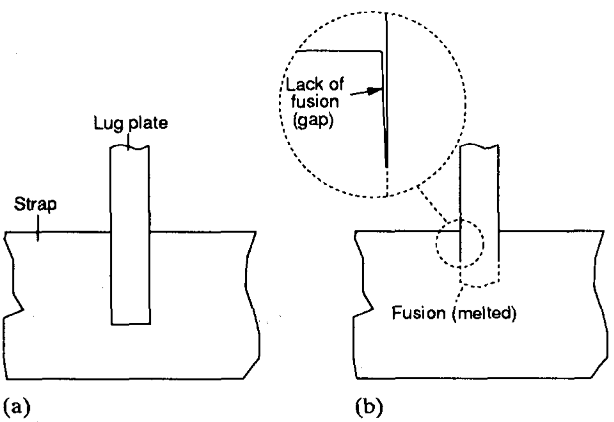


Fig. 6. Schematic of lug after COS operation: (a) no fusion; (b) from lack of fusion to complete fusion.

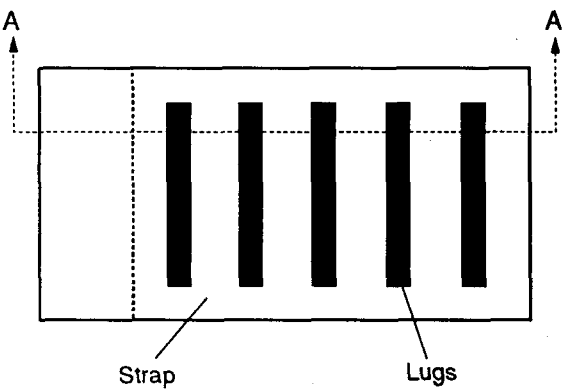


Fig. 7. Schematic illustration of position sectioned (A–A) for metallographic examination. Side view of the joint is shown schematically in Fig. 2.

At the vicinity of the lug/strap interface, pores are often observed. Presumably, these are due largely to the entrapment of gases evolved from the fluxing reaction. Hence, the number of pores was also recorded as an index of joint quality. Figure 6(b) shows that a certain degree of fusion and melting of the lug tip is inevitable for joining of the lug/strap couple. The extent of melting of the lug tip has also been measured and recorded.

For metallographic examination and assessment of a joint, each joint was cold moulded using Struers Epofix. The latter takes 24 h to set. The joints were mounted and sectioned as shown in Fig. 7. The cross section of the joints (indicated by 'A–A' in the Fig.) was prepared for the examination of joint quality.

Results and discussion

Thermal data (no dipping)

Preliminary data using S20 alloy (1.6 wt.% Sb)

Preliminary thermal data using alloy S20 as strap alloy are given in Fig. 8. Figures 8(a) and (b) are thermal cycles for melt pouring temperatures of 500 and 550 °C, respectively, with mould temperatures of 150, 200 and 250 °C. A more detailed graph of the initial cooling stage is given in Fig. 1(c).

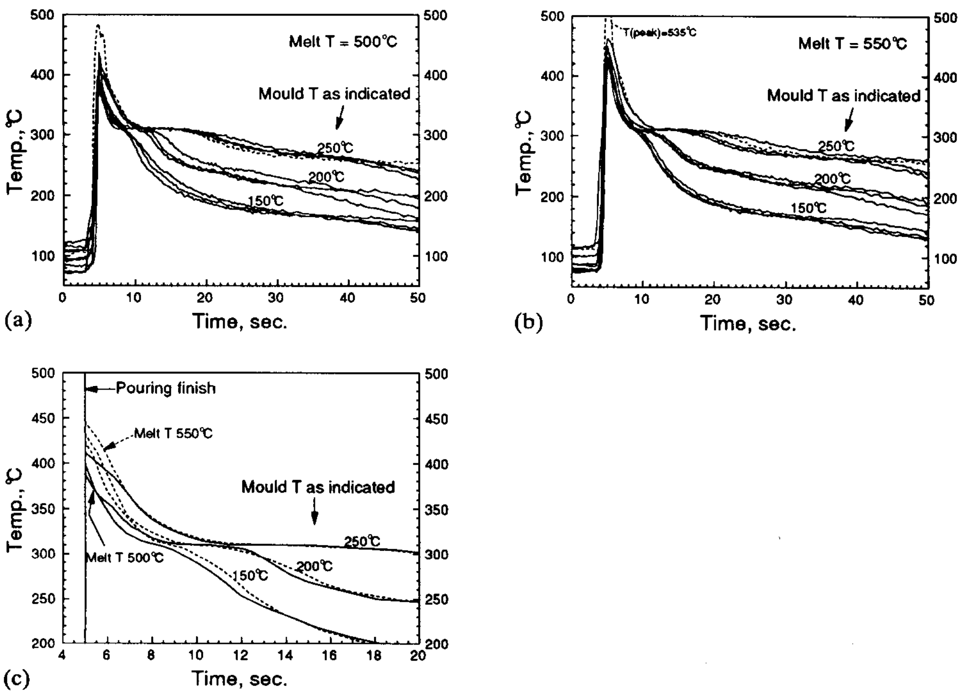


Fig. 8. Thermal cycles of S20 strap samples for melt pouring and mould temperatures as indicated. Broken line in (a) and (b) is for melt that, when poured, first contacts the thermocouple before the wall of the strap cavity. (a) Thermal cycles for melt pouring temperature of 500 °C; (b) thermal cycles for melt pouring temperature of 550 °C; (c) initial cooling curves, each curve is average of three measurements.

During the COS trial, water was turned on to cool the mould when the strap temperature was close to, or less than, the mould temperature. The time at which the water was applied was not controlled and, hence, towards the end of the curves in Figs. 8(a) and (b) there are obvious variations, even for curves for the same mould temperature. The variations in these parts of the curves are not significant when features of the cooling curves are discussed. When the cooling water was not turned on, the strap temperature gradually fell to the mould temperature and became essentially constant towards the end of the 50-s period.

The preliminary data in Fig. 8 show three major features:

(i) for a given melt-pouring temperatures, the mould temperature largely determines the cooling rate (Figs. 8(a) and (b));

(ii) very little influence on the overall cooling rate is observed when the melt-pouring temperature is increased from 500 to 550 °C (Fig. 8(c));

(iii) there is very little variation in the overall measured cooling rate, whether or not the melt, when poured, contacts the thermocouple before the wall of the strap cavity.

A detailed discussion of the features of the cooling curves is given below as part of an analysis of the results obtained for the S31 alloy.

Strap alloy S31 (2.7 wt.% Sb)

Typical thermal characteristics of the commonly used strap alloy (S31) for three mould temperatures are shown in Fig. 9(a). The changes in initial temperature are presented in more detail in Fig. 9(b). The thermal data were obtained for melts that, when poured, contacted the thermocouple first before the wall of the strap cavity. The following three features are observed for the cooling of this strap alloy:

(i) Initial rapid cooling: immediately after pouring, the initial cooling of the strap melt is extremely rapid. The cooling rate is only marginally less for higher mould temperatures as the temperature drops towards the first solidification point (~300 °C).

(ii) First thermal arrest: cooling almost stops, as latent heat is released, when the strap melt starts to solidify at ~300 °C. The duration of this thermal arrest increases considerably with increasing mould temperature.

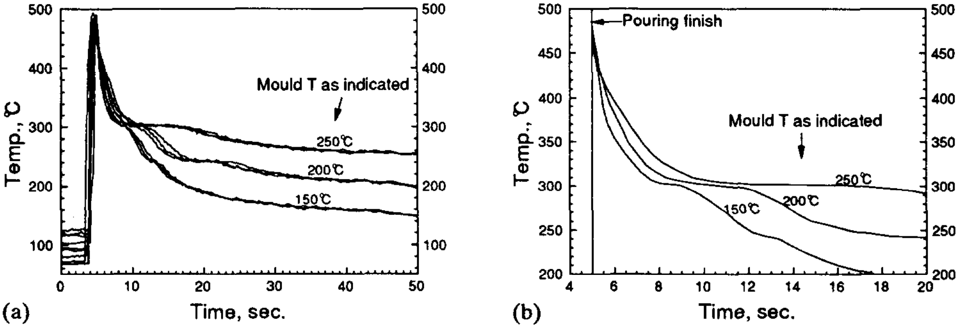


Fig. 9. Thermal histories for S31 strap samples without dipping the lugs at the indicated strap mould temperatures. (a) Thermal histories, three measurements were made for each mould temperature; (b) initial cooling curves, each curve is average of three measurements.

(iii) Second thermal arrest: this thermal arrest is observed at ~250 °C and corresponds to solidification of the eutectic. Once again, higher mould temperatures extend considerably the duration of this thermal arrest.

The initial rapid cooling occurs because of the large differences in the melt and mould temperatures and the high thermal conductivity of the steel mould. The high thermal conductivity and thermal mass of the steel mould also result in there being little variation in the overall cooling curve when the pouring temperature is increased from 500 to 550 °C for the S20 alloy, as noted previously. As the strap temperature decreases towards the mould temperature, the cooling rate decreases because of the diminishing temperature difference.

The cooling curves in the initial rapid cooling region suggest that, for a mould temperature of 150 °C, which is used in current industrial practice, the strap temperature is fast approaching the first solidification temperature (~300 °C) when the much cooler lugs are dipped into the strap. It should be noted that the temperature profiles obtained are for a thermocouple positioned in the middle of the mould cavity. At positions closer to the mould wall, the cooling is expected to be more rapid.

The rate of initial cooling is only marginally less for higher mould temperatures. As shown in Fig. 9(b), however, the temperature of the strap is higher for higher mould temperatures when the lugs are being dipped into the strap. This may be significant for wetting of the lugs during the COS operation.

The two thermal arrests during cooling are due to the evolution of latent heat for the solidification of pro-eutectic and eutectic phases, respectively. Latent heat is released in the entire solidification range from ~300 to ~250 °C. The arrests shown in the cooling curves are the combination of heating from latent heat and cooling by the mould surroundings. The duration of the two thermal arrests increases considerably with every 50 °C increase in the mould temperature from 150 °C. The duration of the second thermal arrest for S20 alloy (Fig. 8) for mould temperatures of 150 and 200 °C is significantly less than that for S31 alloy (Fig. 9). This is expected as there is significantly less eutectic phase formed during solidification of the S20 alloy due to the lower antimony content.

Although no thermal measurements have been made for a real lug/strap couple, the cooling characteristic just described indicate that the heat provided by the cooling strap for joining of the lug/strap couple increases significantly when the mould temperature is increased.

Low-antimony alloy joints without pretinning

Typical lug/strap joint microstructures for low-antimony grid lugs are presented in Figs. 10 and 11 for joints made with mould temperatures of 150 and 200 °C, respectively. During preparation, the samples were ground to about 1.5 mm from the lug edges (Fig. 7). The assessment of joints in terms of the depth of immersed lug, lack of fusion and number of pores is given in Figs. 12 and 13.

Joints made with mould temperature of 150 °C

For joints made at a mould temperature of 150 °C, the joints can be characterized by the gradual increase in the degree of fusion from the strap surface to the lug tip. As shown in Fig. 10, the lug/strap couple starts with a complete lack of fusion at the strap surface and leaves an obvious gap between the couple. The gap width decreases towards the lug tip. The gap starts to close completely when partial fusion is observed. This can be regarded as the start of the true joint for the lug/strap couple. The partial fusion results from the melting of the low-melting-point eutectic on the lug surface

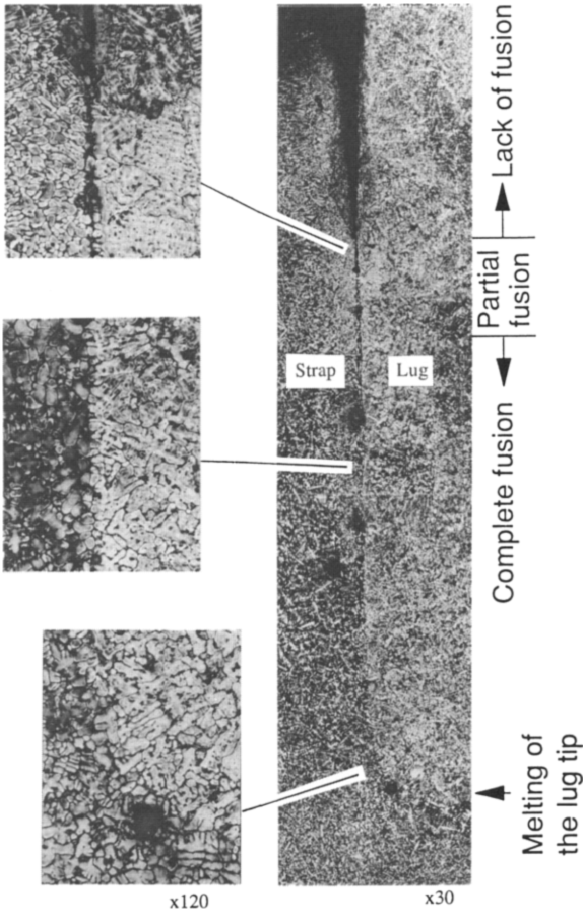


Fig. 10. Microstructure of lug/strap joint using low-antimony alloy lug with mould temperature of 150 °C.

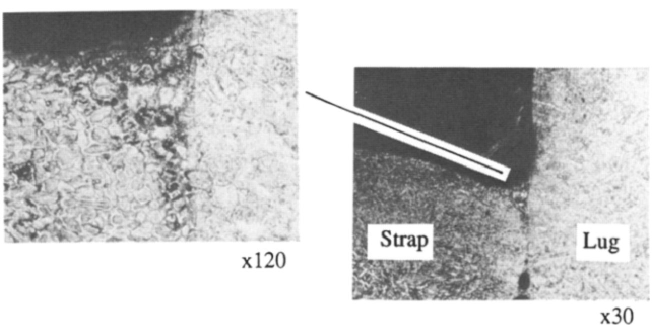


Fig. 11. Microstructure of lug/strap joint using low-antimony alloy lug with a mould temperature set at 200 °C.

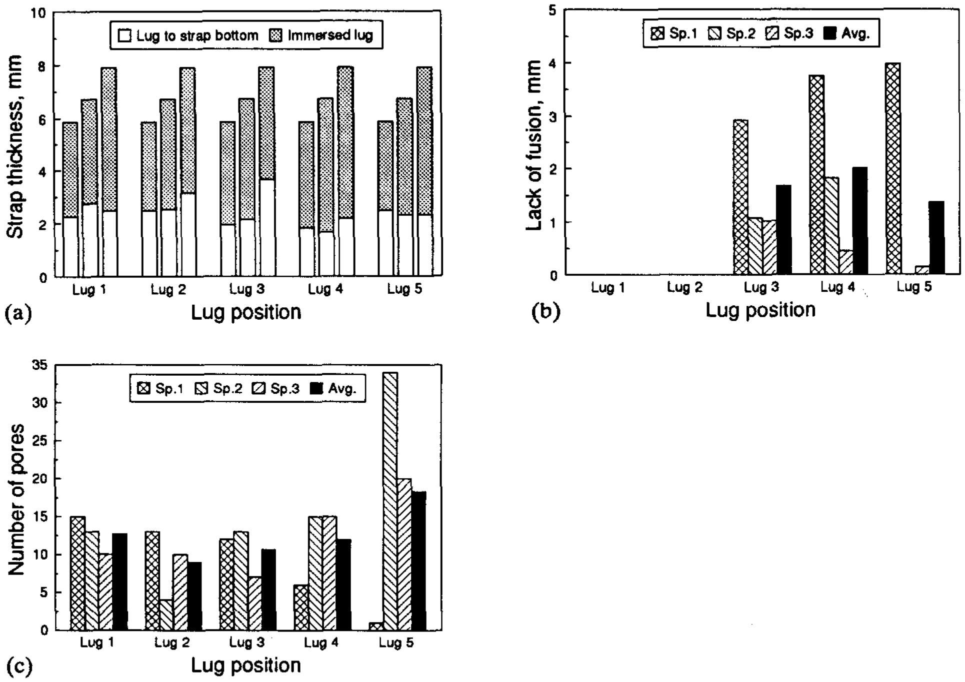


Fig. 12. Quality assessment of joints made using low-antimony alloy lugs (no tinning) with mould temperature of 150 °C. Sp. 1 refers to thinnest strap of the three, etc. (a) Strap thickness; (b) lack of fusion; (c) number of pores.

and mixing of this eutectic with the strap melt during joining. Towards the lug tip, the lug surface was completely melted during joining; this is shown in the micrographs in Fig. 10.

Although no temperature measurements were made for the lug/strap joint, the nature and degree of fusion results from thermal changes during dipping of the lug into the strap melt. As a lug is dipped into the strap melt, heat is transferred from the strap to the lug. Positions closer to the lug tip were immersed longer in the strap melt and hence achieved a higher degree of fusion. With respect to the strap, the positions closer to the melt surface were cooler due to a greater loss of heat during dipping. Hence, this region of the melt provides less heat for fusion with the solid lug in the later stages.

The micrographs in Fig. 10 also show that there are a number of pores at, or near, the lug/strap interface. These pores presumably result from the entrapment of gases evolved from the fluxing reaction.

The quality assessment of the three joints made for a mould temperature of 150 °C is given in Fig. 12. In Fig. 12(a), the average thicknesses of the three straps (same in each lug position) are given together with the measured lengths of the lugs immersed in the straps. The distance between the lug tips to the strap bottom was set at slightly less than 2 mm. Figure 12(a) indicates that some lug tips were slightly melted off during the COS joining as some lug tips are more than 2 mm away from the strap bottom. This melting of the lug tip is indicated in the micrographs of Fig. 10.

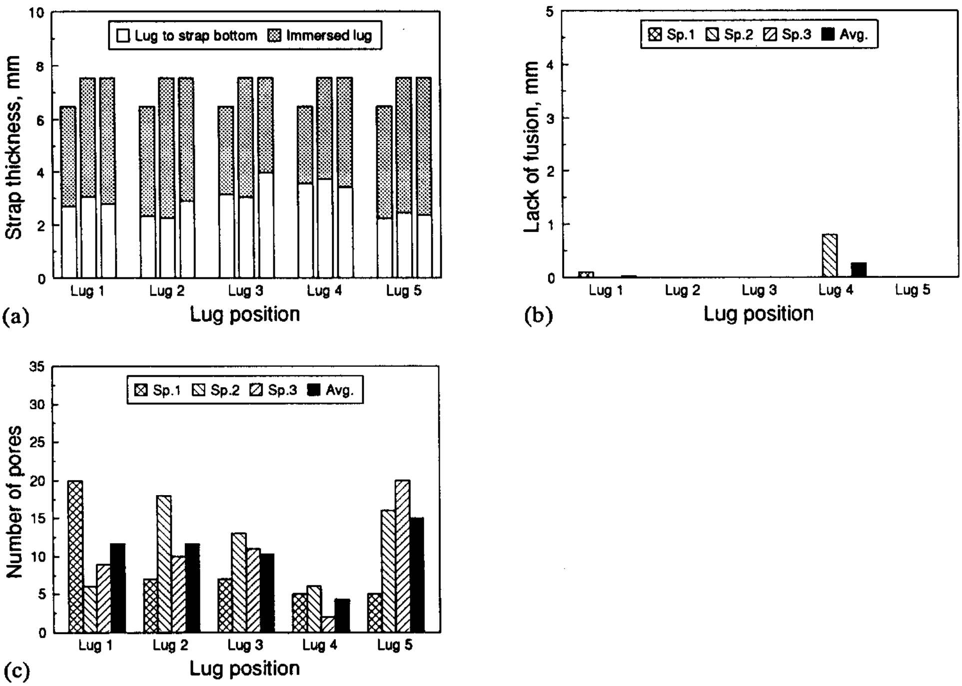


Fig. 13. Quality assessment of joints made using low-antimony alloy lugs (no tinning) with mould temperature of 200 °C. Sp. 1 refers to thinnest strap of the three, etc. (a) Strap thickness; (b) lack of fusion; (c) number of pores.

Figure 12(b) shows that a significant lack of fusion occurs with some of the lugs. This lack of fusion is not symmetrical and is probably due to the presence of the vertical portion of the mould cavity that is used to simulate the necessary part for through-the-partition joining. For lugs closer to this vertical portion, cooling is expected to be less rapid as the heat loss to the mould is slower.

A point to be noted in Figs. 12(a) and (b) is that sample 1 (the left bar given for each lug) has the thinnest strap and highest degree of lack of fusion. This indicates that with a smaller amount of strap melt, the rate of heat loss is higher and results in greater lack of fusion.

Figure 12(c) shows that a high level of porosity is present in the joints. In lug 5 (Figs. 12(b) and (c)), samples 2 and 3 each show relatively good fusion, but a high number of pores. By contrast, sample 1 exhibits the opposite behaviour. This is probably because the presence of a gap, due to very poor fusion, allows gases to escape more easily.

From the above discussion, it may be concluded that, for a mould temperature of 150 °C, cooling of the strap melt is too rapid to prevent the occurrence of very poor fusion or a high level of porosity. This confirms Prengaman's [1] observation that joining of the lugs to straps is a major cause of manufacturing defects in batteries.

Joints made with mould temperature of 200 °C

Increasing the mould temperature to 200 °C significantly increases the degree of fusion as shown in Fig. 13(b). This can also be concluded by examination of micrographs

(Fig. 11) that shows that the lug joins the strap to the strap surface. This better joining results directly from the increase in the heat provided by the cooling strap for joining when the mould temperature is increased. The extent of melting of the lug tips is also seen to increase slightly, cf., Fig. 13(a) with Fig. 12(a). Nevertheless, the number of pores remains high (Fig. 13(c)).

Both simple thermal characteristics, as discussed previously, and examination of the joints suggest that mould temperature (T_M), or the temperature difference ($\Delta T_{S\text{-}M}$) between the solidification temperature (T_S) and the mould temperature, is a very important parameter that affects joint quality. Increasing T_M, or decreasing $\Delta T_{S\text{-}M}$, provides more heat and more time for the strap to more completely wet the lugs. This gives more melting of the lugs and improves joint quality.

Low-antimony alloy joints with pretinning of lugs

Typical joint microstructures using pretinned lugs are shown in Fig. 14. The results of the quality assessment are given in Fig. 15 for a mould temperature of 150 °C only. The joints made with pretinned lugs show excellent joining without any lack of fusion and, in this respect, differ markedly from those without pretinning. There are also fewer pores when pretinned lugs are used.

The excellent joining achieved with pretinned lugs, even at a mould temperature of 150 °C, is because of filling of the gap by tin or tin–lead mixture, as indicated in Fig. 14. As tin melts at a significantly lower temperature (232 °C) than the strap melt (at least 300 °C), the tin that is coated on the lugs melts easily whilst the lugs are being dipped into the strap. This ensures 'true' wetting between the lugs and the

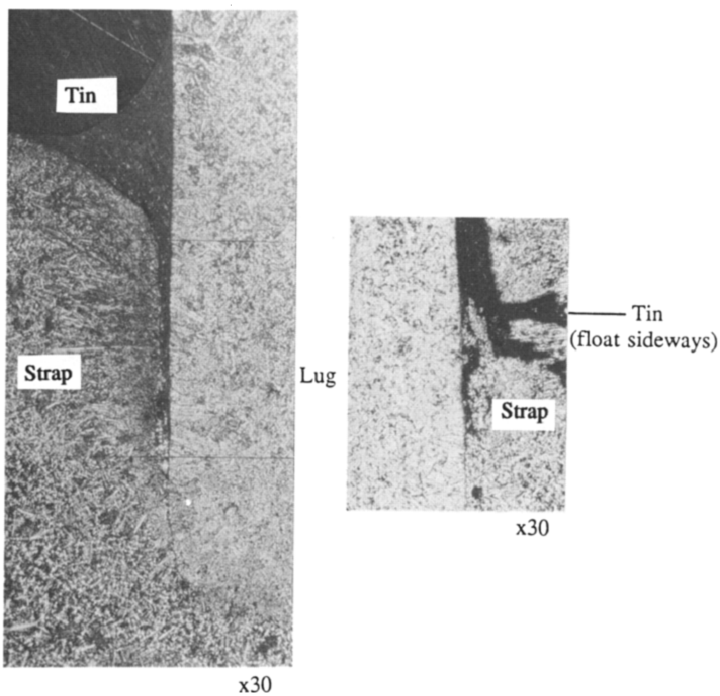


Fig. 14. Microstructure of lug/strap joint using pretinned low-antimony alloy lug with mould temperature set at 150 °C.

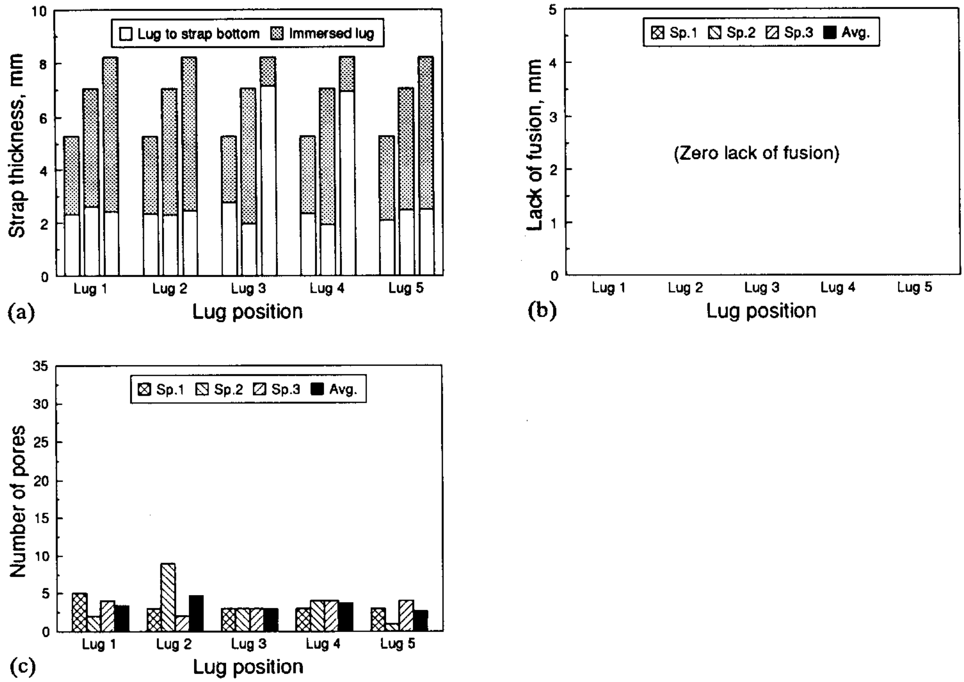


Fig. 15. Quality assessment of joints made using pretinned low-antimony alloy lugs with mould temperature 150 °C. Sp. 1 refers to thinnest strap of the three, etc. (a) Strap thickness; (b) lack of fusion; (c) number of pores.

strap. Tin also melts easily and alloys with the strap and, thus, the material in the region between the lugs and strap has a lower melting point (or melting range). The melt is liquid for longer in this region and gases from the fluxing reaction also escape more easily and give less porosity.

Tin has a much lower density than lead and, hence, will tend to float on the top of the molten alloy in the strap cavity. Molten tin can also float off sideways from the lug (Fig. 14). This may result from the combination of the violent fluxing reaction and melting of the thick tin coatings (particularly the drip on the lug tip). Rapid freezing of the strap melt prevents further alloying or floating of the molten tin on the surface of the strap.

Lead–calcium

Figures 16 and 17 give typical microstructures and quality assessments for joints made using lead–calcium alloy lugs with a mould temperature of 150 °C only. These data show that complete joining of the lugs and the strap without lack of fusion is achieved, and that the porosity level is low for the calcium alloy joints.

Different fluxes were used for the low-antimony alloy lugs and the calcium lugs. Hence, these joints cannot be simply compared. The flux (TBS no. 6) used for the calcium alloy lugs is acid-based and has a more aggressive fluxing action than the oil-based flux (TBS no. 5) that was used for low-antimony alloy lugs [9]. Thus, no. 6 flux promotes true wetting and gives sound joints. The low level of porosity for the calcium alloy joints occurs possibly because the gases generated from fluxing are less when no. 6 flux is used compared with no. 5 flux.

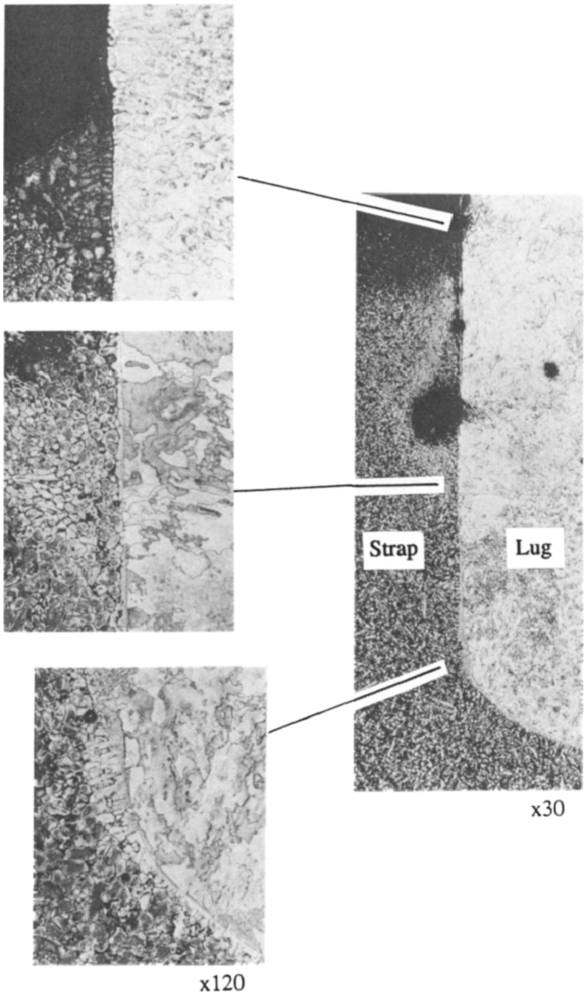


Fig. 16. Microstructures of lug/strap joint using calcium alloy lug with mould temperature of 150 °C.

As shown in Fig. 16, joining of the calcium lugs and the strap is characterized by the growth of pro-eutectic (lead–antimony) phase from the original grains in the calcium lugs. This resembles solidification during welding in which the weld metal begins to solidify from the grains of the parent metal. This feature of the interface microstructure is the direct result of the fusion of the lug surface during the COS process, and is analogous to melting of the parent metal during welding by the welding arc. This structure is more apparent for lead–calcium alloy as the microstructure of the lug is different from that of the strap alloy.

Conclusions

1. Initial cooling of the strap melt is extremely rapid immediately after pouring. Increasing mould temperature in the range from 150 to 250 °C only marginally reduces

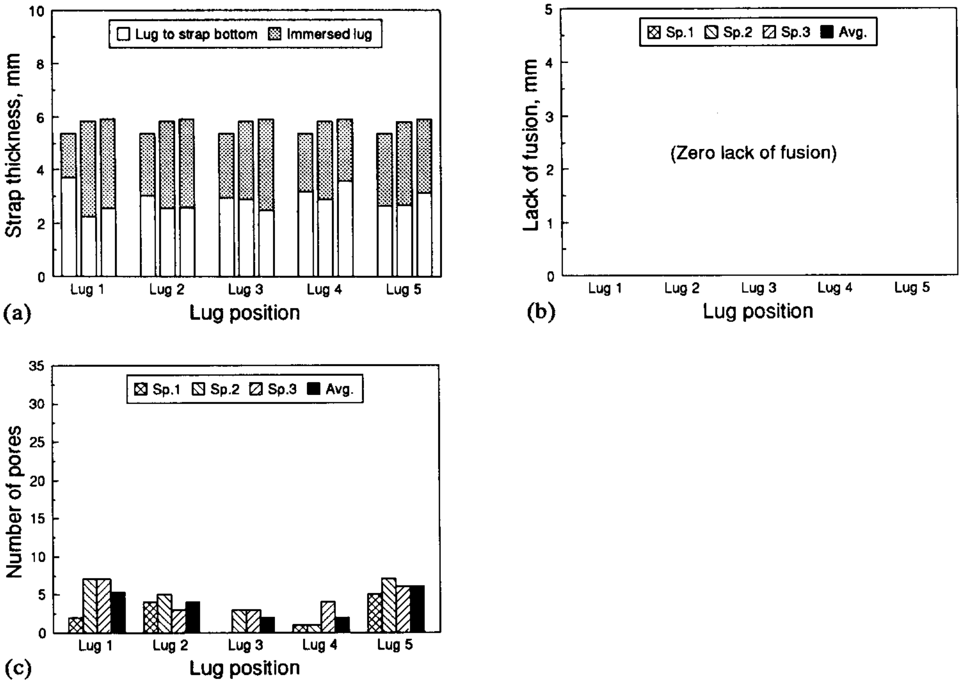


Fig. 17. Quality assessment of joints made using calcium alloy lugs with mould temperature, 150 °C. Sp. 1 refers to thinnest strap of the three, etc. (a) Strap thickness; (b) lack of fusion; (c) number of pores.

the initial cooling rate, but lowers considerably the subsequent cooling rate when the strap melt starts to solidify. Increasing the melt-pouring temperature from 500 to 550 °C has little influence on the overall cooling characteristics of the strap.

2. Joining of the lug/strap couple can be characterized by lack of fusion, partial fusion and complete fusion from the strap surface to the lug tip. Complete fusion of the strap and lugs is characterized by the growth of pro-eutectic phase (lead–antimony) in the strap from the original grains in the lugs.

3. At a mould temperature of 150 °C, a significant lack of fusion and considerable porosity are observed for low-antimony alloy joints without pretinning of lugs. The degree of fusion increases significantly when the mould temperature is increased to 200 °C.

4. Pretinning effectively eliminates lack of fusion and considerably reduces the level of porosity. The beneficial effect of pretinning is most likely due to the presence of lower melting point material between the lugs and the strap. This material promotes wetting, melting and joining by prolonging the melt life in the interface region and bridging the lugs and strap.

5. There is no lack of fusion and the level of porosity is low for lead–calcium alloy joints for which a more aggressive acid-based flux is used.

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