

STUDY OF LIQUID FILM MIGRATION IN O-TEMPER AA3005 BRAZING SHEET

Aad J. Wittebrood*, Rinze Benedictus*, Klaus Vieregge**

* Hoogovens Research & Development
P.O. Box 10000, 1970 CA, IJmuiden
The Netherlands

** Hoogovens Aluminium Walzprodukte GmbH
Carl-Spaeter-Strasse 10, D-56070 Koblenz
Germany

ABSTRACT A study has been carried out to determine the nature of core penetration sometimes observed in O-temper brazing sheet. It became clear that the grain size of the core plays an important role. From the results, the Liquid Film Migration as described by the coherency strain theory could not be fully corroborated. According to this theory the stress in the aluminium matrix caused by the amount of manganese in solid solution is a necessary condition for LFM. According to the results presented here, identical concentrations of Manganese in solid solution can give substantially different LFM. The investigation can not yet give a definite answer as to why samples with a certain amount of cold work show after recrystallisation no LFM of the core. It is however evident that the distribution, size and volume fraction of manganese bearing precipitates play an important role

Keywords: *Brazing Sheet, Erosion, Grain Boundary, Precipitates, Liquid Film Migration*

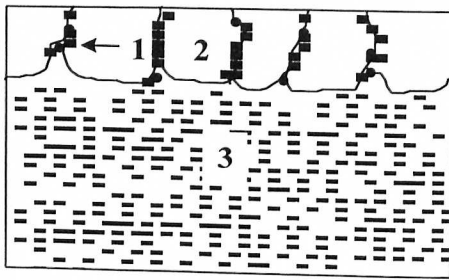
1. INTRODUCTION

Aluminium Brazing Sheet is used for the industrial production of heat exchangers for the automotive industry. Brazing sheet is typically a two layer aluminium system, i.e. a core alloy responsible for giving the final product strength and corrosion resistance, and a clad alloy responsible for supplying the material for joining. In most cases the core alloy is an AA 3XXX type alloy and the clad alloy an AA 4XXX alloy.

Due to forming operations necessary to obtain the shape needed for a specific heat-exchanger design, the brazing sheet is often supplied in O temper condition. During brazing the 4XXX alloy with a liquidus temperature below that of the 3XXX alloy will melt, and through capillary- and surface tension forces will close the gaps between the components of a heat exchanger, resulting in a complete, one step joined product. During this brazing process changes take place at the interface between the clad- and core alloy. In the case of the O temper condition, severe penetration of the clad alloy into the core alloy can take place. This penetration results in less flow of the clad alloy giving smaller joints. Together with this reduced flow, the penetration leads to erosion of the core alloy, inducing metallurgical and micro-structural changes, giving the final product less strength and corrosion resistance.

Figure 1 is a schematic presentation of brazing sheet after brazing suffering from LFM. Under the residual clad layer, a layer is formed with a different chemical composition compared to the original core alloy. The chemical changes have been well investigated by Woods[1]. Several early papers[2,3,4] have been published describing this phenomenon, but all give a different explanation for the mechanism behind this phenomenon. Later[5] it was described that the erosion was dependant on the number and size of the precipitates in the core alloy. Only recently first attempts were made to explain the phenomenon thermodynamically[6]. All these publications dealt with brazing sheet material.

Figure 1 Schematic representation of the altered structure after brazing



- 1- α -Al(FeMn)Si particles
 2- dispersoid free material, alpha aluminium
 3- Original core alloy

In 1997, Woods[1] and Yang[7] were the first to relate the observed phenomenon in brazing sheet with Liquid Film Migration (LFM). LFM has been a topic of research since the seventies. Yoon[8] wrote an excellent overview presenting a theory of explaining LFM. This theory is called the diffusional coherency strain theory. The

observed changes can partly be related to this theory but still some questions remain unanswered. This paper tries to relate the observations with this theory, but will raise questions on some of the statements in earlier publications.

2. MATERIALS AND METHODS

2.1 Materials

For the study, an AA3005 alloy cladded with an AA4047 alloy was used. Table 1 shows the maximum concentration of the different elements in both alloys.

Table 1 Alloy chemistries

| Alloy | Mn | Si | Fe | Mg | Cu | Cr | Zn |
|--------|------|------|------|------|------|------|------|
| AA3005 | 1.50 | 0.60 | 0.70 | 0.60 | 0.30 | 0.10 | 0.25 |
| AA4047 | 0.15 | 13.0 | 0.80 | 0.10 | 0.30 | - | 0.20 |

Sample blocks from the core alloy (AA3005) were given different heat treatments before they were cladded with the AA4047 alloy. Heat treatments are shown in table 2.

Heat up and cooling rates were the same for all blocks.

Table 2 Heat treatments

| Code | Temperature (°C) | Time (hrs) |
|------|------------------|------------|
| B4 | - | - |
| G3 | 500 | 10 |
| E2 | 550 | 10 |
| G2 | 575 | 10 |
| A2 | 600 | 4 |
| B2 | 600 | 10 |
| C2 | 600 | 24 |

Hot rolling, cold rolling and inter-annealing conditions were the same for all samples. To investigate the effect of the grain-size, the material was inter-annealed at different gauges (A and B). (gauge A > gauge B) The end-anneal temperature was 350 °C or 375 °C.

The thickness of the brazing sheet used for the experiments was 0.50 mm, the clad was 10 % of the total thickness.

To study the effect of pre-stretching, variant B2 was processed differently with respect to the position of the inter-anneal. This was

carried out to assure a different grain size in the end-annealed condition. The material was stretched on a tensile machine for fixed percentages. Brazing took place in a vacuum brazing furnace with a maximum hold temperature of 600°C for 3 minutes.

2.2 Experimental Methods

Brazeability or fillet sizes were evaluated with a T-sample. Determination of the fillet area was done as described earlier[9]. The areas measured were expressed as a percentage of the total clad area available for fillet forming. In the measured samples the total area for fillet formation is: $((\text{clad thickness})50 \times 10^{-6}) \times ((\text{T-sample width})12 \times 10^{-3}) = 6.0 \times 10^{-7} \text{ m}^2$

The same samples were used to determine the depth of penetration.

Manganese in solid solution was determined by measuring the conductivity. All samples were measured after annealing or brazing.

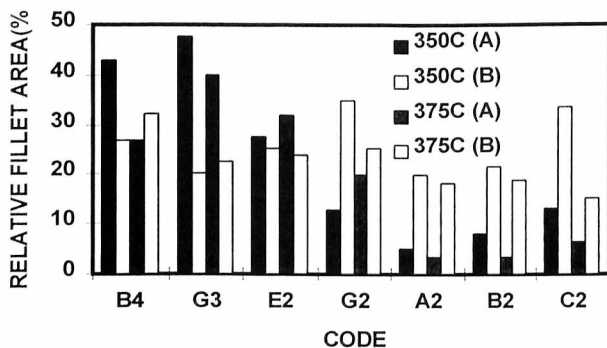
Grain size measurement, the samples were polished, etched with Barker's reagent and anodised. ASTM grain size measurement was performed according to E 112.

3. RESULTS

Effect of heat treatment

Figure 2 shows the sample codes corresponding to the different heat treatment against measured fillet sizes. The fillet sizes are given for the samples having a different point of inter-anneal together with a different end-anneal temperature.

Figure 2 Relative fillet sizes for different heat treatments and end-annealing temperatures



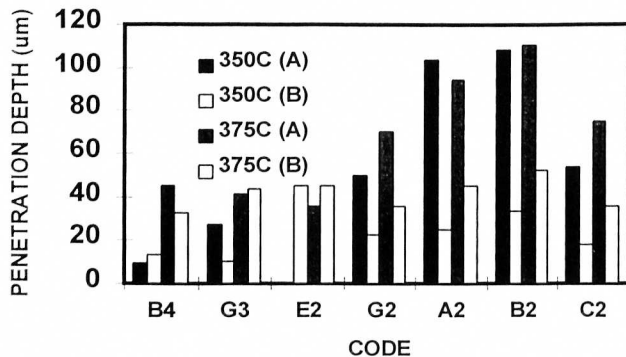
The legends correspond with the different end-anneal temperatures and between brackets the different inter-anneal gauges

Table 3 Grain sizes for different heat treatments

| Code | E.A. 350°C | | E.A. 375°C | |
|------|------------|--------|------------|--------|
| | I.A. A | I.A. B | I.A. A | I.A. B |
| B4 | * | * | * | * |
| G3 | 7.0 | * | 7.1 | 6.9 |
| E2 | 7.6 | 7.5 | 7.6 | 7.7 |
| G2 | 8.4 | 7.5 | 8.1 | 7.5 |
| A2 | 8.1 | 7.1 | 8.6 | 7.5 |
| B2 | 8.7 | 7.6 | 8.4 | 7.5 |
| C2 | 8.3 | 7.2 | 8.2 | 7.4 |

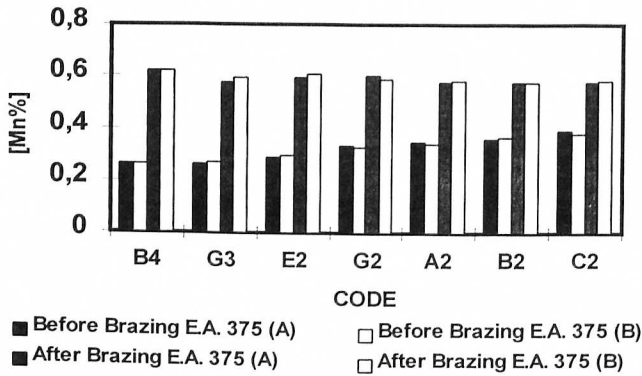
* Not fully recrystallised

Figure 3 Depth of penetration after brazing



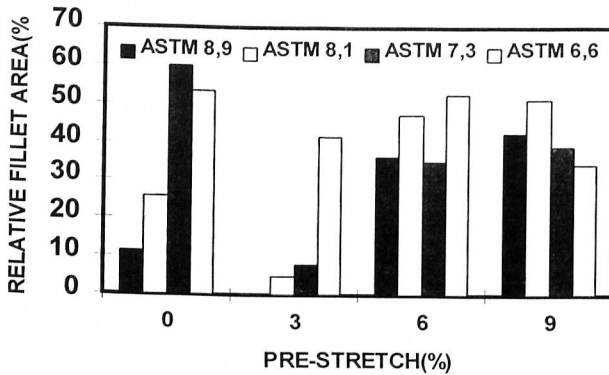
The legends correspond with the different end-anneal temperatures and between brackets the different inter-anneal gauges.

Figure 4 Mn in solid solution during processing, before and after brazing



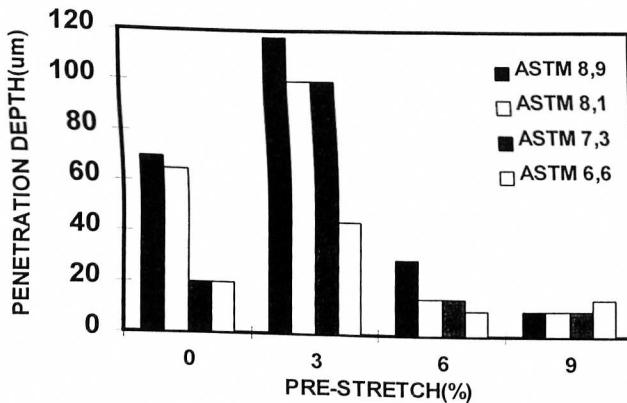
The legends correspond with the different end-anneal temperatures and between brackets different inter-anneal gauges.

Figure 5 Relative fillet areas for different grain-sizes and pre-stretch conditions



Processing took place according to sample B2

Figure 6 Depth of penetration after brazing for different grain-sizes and pre-stretch



4. DISCUSSION

Figure 7 shows that there is a relation between fillet size and penetration depth. Deep penetration gives a small fillet. The formation of the so-called liquid film migration zone extracts silicon from the clad layer. In this way the melt temperature of the filler is increased thereby decreasing the time

of the liquid state of the filler during the braze process, and reducing the amount of available filler alloy. This results in less filler flow and smaller fillets. Understanding why the penetration is occurring is essential to understand since the future trend is to use thinner material and less clad alloy.

Figure 7 Relation between penetration depth and fillet size

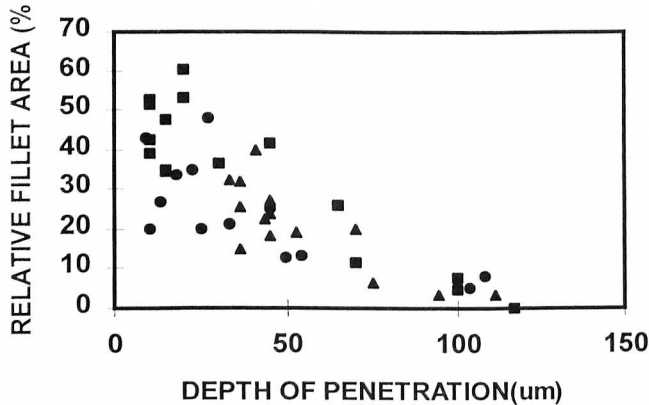


Figure 7 contains all data points from this study.

Effect of heat treatment

On first sight, the lowest heat treatment temperatures give the best results, but as indicated in table 3 these samples were not fully recrystallised. As a result from grain boundary pinning caused by small precipitates formed at the low temperature, samples B4,G3, E2 showed long elongated grains. The recrystallised samples (G2,A2,B2 and C2) with the inter-anneal at gauge B showed the largest fillet area. Through less cold work prior to end -annealing, larger grains in the final condition can be obtained. Materials with larger grains show less penetration. It can thus be concluded that the amount of grain boundaries play a role in the observed penetration. Grain boundary diffusion seems to be a prerequisite for penetration. In a brazing sheet system, melting starts preferentially at the clad/core interface. Melting starts, some degrees lower than the eutectic temperature of 577 °C because of the copper, magnesium and manganese in the core. According to Nylén[6] the liquid first wets the grain boundaries, then the liquid film is dragged into the core due to the matrix grain boundary tensions. The liquid film does not solidify according to Nylén[6] because of the dissolution of the second phase particles. This lowers the melting temperature.

According to Woods[1] and Yang[7] the energy to keep the film molten is a result from the coherent strain energy. In this, grains are not in chemical equilibrium with the liquid in which they are imbedded. The system tries to establish a new equilibrium at the interfaces by diffusion from the grains into the liquid phase or visa versa. This leads to a coherently strained surface layer on the grains. If the two liquid-grain interfaces are not identical, there will be an anisotropy in the strain energy. This will direct the movement of the liquid film. According to Yoon[8] the energy of mixing keeps the film molten and the strain only dictates the direction of movement.

Material heat treated at 600°C for 4 and 10 hours showed the largest fillets especially when the inter-anneal was carried out at the lowest inter-anneal gauge(A). Why these samples have the best brazing property is difficult to explain with the coherency strain theory as interpreted by Woods[1] and Yang[7]. According to them the amount of manganese in solid solution is an important parameter. The stress relief during brazing is the energy source that controls the movement of the layer. If one compares the different concentrations of manganese in solid solution for the different samples, the difference in brazing can not be explained by this concentration before or after brazing.

During heat treatment precipitates are formed. These precipitates can either act as nucleation sites for new grains or when they are small, can pin grain boundary movement during recrystallisation. In the last case, if the liquid filler penetrates the core along the grain boundaries, the melting point of the filler will drop through the dissolution of the precipitates causing more penetration. When the precipitates are large enough, they will not be dissolved into the liquid filler and thus less penetration will occur.

Effect of pre-stretching and grain sizes

At 3% stretching severe core penetration is observed. The material does not recrystallise before brazing. Penetration will occur through grain boundaries and dislocation structures. In the literature[5,7] no explanation is given why recrystallisation prevents penetration of the core. In the case of low reductions, recrystallisation takes place at a high temperature. During the brazing process silicon diffuses along the fast diffusion paths like grain boundaries and dislocation structures. Recrystallisation rearranges the micro-structure, changing the diffusion paths. To meet the requirement for LFM, silicon has to start diffusing into the new micro-structure to reach the necessary level to start LFM.

The explanation above is not valid for material that recrystallises at lower temperature after higher deformation energy. In this case, there is enough time for the silicon to diffuse into the core and to offset LFM. The distribution (located on grain boundaries or in the grain itself) and size of precipitates in the core alloy will have an impact on the occurrence of LFM as stated by Yamauchi[5].

5. CONCLUSIONS

Large grains give less LFM, due to slower diffusion paths. With comparable grain sizes the place where the manganese precipitates are present is important. Recrystallisation changes the position of the precipitates relative to the grain boundaries, in this way affecting the susceptibility of the core alloy to LFM.

Just based on the amount of manganese in solid solution before and after brazing one can not predict that LFM will happen. If the coherency strain energy is coming from the change of manganese in solid solution, then the occurrence of LFM as observed in brazing sheet can not be explained by the coherency strain theory.

The model proposed by Nylen[6] is a better explanation for the phenomenon observed. Especially when one considers the position of the precipitates that pin grain boundaries.

Experiments are being set-up to investigate the influence of the time involved with the brazing process and if this can offset LFM in pre-stretch material.

6. REFERENCES

- [1] R.A. Woods, SAE 971848, VTMS 3 Conference, 19-21 May 1997, Indianapolis, USA
- [2] Sharples, Weld. J., 54 (1975), 164
- [3] D.J.Schmatz, Welding Research Supplement, October 1983, 267-271
- [4] Ikuo Okamoto, Tadashi Takemoto, Kei Uchikawa., Transactions of JWRI, vol. 12, no. 1, 1983, 57-64
- [5] Shigenori Yamauchi, Kenji Kato, The Proceedings of International Conference on Recent Advances and Engineering of Light Metals, October 1991, Tokyo, Japan.
- [6] Nylén, U. Gustavsson, B. Hutchinson, A. Örtén, Materials Science Forum Vols. 217-22(1996) pp. 1703-1708
- [7] H.S. Yang, R.A. Woods, SAE 971849, VTMS 3 Conference, 19-21 May 1997, Indianapolis, USA
- [8] D.Y. Yoon, International Materials Review. 1995, vol 40, no.4, pp149-179
- [9] A.Wittebrood, T. van der Veldt, K. Vieregge, A. Haszler, SAE 971858, VTMS 3 Conference, 19-21 May 1997, Indianapolis, USA