Effects of Surface Preparation on the Microstructure of ARB Processed Aluminium

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Abstract

Sheets of commercial purity aluminium have been deformed by Accumulative Roll-Bonding (ARB), utilising two different techniques to prepare the bonding surfaces prior to rolling; namely wire brushing and a hot caustic etch. The effects of these preparation methods on bonding, microstructure and the oxide particles at the bond interface have been examined using optical microscopy, a FEG-SEM, and TEM. Wire brushing was found to produce a much rougher surface than caustic etching and gave a more consolidated bond during ARB processing. However, it was also found to introduce high levels of deformation into the surface of the sheet and TEM analysis revealed this produced bands of nano-scale grains at the interface between bonded layers, resulting in a heterogeneous microstructure through the sheet thickness. TEM analysis revealed evidence of aluminium oxide becoming incorporated into the bulk of the ARB processed sheets.

1. Introduction

It has been known for some time that materials with ultra-fine grain (UFG) structures exhibit advantageous properties, including increased strength and high strain rate superplasticity [1]. Severe deformation processing has recently become of increasing interest due to its capacity for producing UFG structures in bulk materials at a relatively low cost [1]. One such severe deformation process, accumulative roll-bonding (ARB), has the attraction that it can potentially be used for producing large sheet materials containing these UFG structures. The ARB process involves roll-bonding two sheets, using a 50% reduction, to a thickness equivalent to that of the original starting sheet. The roll-bonded sheet is then cut in half and the two halves are stacked on top of one another and the process repeated until the desired strain is achieved. Theoretically, the technique can be used to build up unlimited strains in a material, because there is no change in the sheet dimensions. A significant amount work has been published relating to the microstructures produced during ARB processing of different materials (e.g. [2-4]), but little has been reported concerning the effects of surface preparation and the bond interface on the sheet structure.

ARB processing [2,3] is different to many other severe deformation techniques in that each cycle involves bonding two sheets together in one roll pass. In order to produce a bulk homogeneous material, a good consolidated bond has to be achieved. In nearly all the work published, relating to ARB processing of aluminium alloys, wire brushing has been used to prepare the surfaces prior to roll-bonding, in order to remove the aluminium oxide layer [3,5].

However, wire brushing will clearly be intrusive to the surface and will add another deformation process to the equation. Additionally, because of the immediate formation of a passivating aluminium oxide layer when the pure aluminium is exposed to the atmosphere, some oxide will undoubtedly be rolled into the sheet. During ARB processing the number of bonded layers doubles with each ARB cycle and after 10 ARB cycles the number of bonded layers will be slightly in excess of 1000. If processing is carried out using 2 mm thick sheets, this will result in each layer being only $\sim 2 \,\mu\text{m}$ in thickness. This means that any heterogeneities in the surface prior to roll-bonding will become included within a large proportion of the sheet. If, for example, a large amount of oxide is rolled into the sheet during each ARB cycle, then after 10 ARB cycles the sheet could effectively become a mechanically alloyed composite of layered aluminium and oxide. Furthermore, wire brushing introduces a large degree of local strain into the sub-surface, and could act as an additional grain refinement mechanism. With the accumulation of layers, and hence surfaces, within the material this could lead to a heterogeneous distribution of grain size throughout the sheet thickness.

This current investigation will examine the degree to which each of the features, mentioned above, affect the ARB processed sheets and their microstructures. The effect of wire brushing on the microstructure at the surface and at the interfaces between layers in ARB processed sheets will be investigated, in addition to that of using chemical preparation methods on the bonding between sheets. Furthermore, TEM studies carried out to investigate the inclusion of aluminium oxide during ARB processing will be discussed.

2. Experimental Procedures

Two separate sets of ARB processing were carried out on 2mm thick sheets of conventionally rolled commercial purity (99.7%) aluminium (AA1100) (with an initial von Mises strain of 4). The first set of ARB processing utilised wire brushing to prepare the bonding surfaces prior to each roll pass. This involved degreasing the surfaces with acetone followed by subsequent wire brushing to remove the aluminium oxide layer. The wire brushed surfaces were then cleaned with acetone before the sheets were stacked and roll-bonded using a 50% reduction. The second set of ARB processing employed a chemical treatment to prepare the bonding surface, imparting no deformation to the sheet surfaces. This involved degreasing with acetone, followed by an etching treatment in a caustic solution (5% NaOH, 4% NaNO₃) for 5 min at 65°C, to remove the aluminium oxide layer. After being rinsed in water, the sheets were desmutted in 25% nitric acid (HNO₃) followed by another water rinse and drying. The wire brushed surfaces were examined prior to and after ARB processing using a Philips XL30 FEG-SEM and a Philips CM200 TEM respectively. Secondary electron (SE) imaging in the FEG-SEM was carried out to enable the surface topography to be examined. TEM was used to explore the microstructure at the interfaces between bonded sheets, using thin foils prepared from the ND-RD plane of ARB processed specimens electropolished at -30°C in a standard nitricmethanol solution. Optical microscopy was also carried out to compare the difference in bonding between wire brushed and chemically etched sheets. TEM analysis was additionally used to investigate the extent to which aluminium oxide is incorporated into the sheets during ARB processing.

3. Results and Discussion

3.1 The Wire Brushed Surface

SE images revealing the surface topography of the wire brushed surface are shown in Figure 1. At low magnifications (Fig1a), the surface appeared fairly heterogeneous, with scratches from the wire brush clearly visible. At higher magnifications other features were revealed, most interesting of which were *hills, troughs* and *overhangs* that covered the surface (Figure 1b). During roll-bonding, these features may interlock with each other and aid bonding between the two sheets. However, flakes of metal were also visible on the wire brushed surface, which if loose, could hinder bonding between the two sheets (Figure 1c), thus causing structural weaknesses along sheet interfaces. The SE images also showed that wire brushing caused regions of material to fold over each other, which may result in other heterogeneities such as dirt, grease or oxide becoming embedded within the sheet surface. Since the number of bond interfaces doubles with each ARB cycle, any heterogeneities caused by the rolled-in surfaces are progressively dispersed throughout the sheet thickness, and could affect grain refinement as well as the resultant behaviour on annealing and the sheet's mechanical properties.



3.2 Microstructure of the Wire Brushed Interface Layers

As well as producing a very rough surface, the high level of local deformation imparted by wire brushing can affect the bond line microstructure [6], which will then become dispersed throughout the sheet in subsequent ARB passes. This behaviour was explored in more detail by using TEM to analyse the microstructure at the interfaces between bonded layers. Figure 2a shows TEM bright field images of a typical interface between two bonded layers, after 7 ARB cycles ($\epsilon = 9.6$). This interface was 45 μ m from the centre bond-line and would have been created during one of the first two ARB cycles. It can be observed that at the bond-line there is region of material, of irregular width (up to ~ 2 μ m thick in places), which appears different to that of the surrounding matrix. The variation in

thickness, despite the large subsequent strain after the bond being formed, is presumably due to the incorporation of folds and flakes into the interface. On closer inspection, it can be seen that in places there was still incomplete bonding, particularly where flakes where incorporated into the surface (arrow in Figure 2b). The interface layer was comprised of lamellar boundaries and 'cells' that were much smaller in size than those in the bulk of the sheet (Figure 2b). The lamellar boundaries were of around 100 –200 nm wide, compared to a cell size of ~ 0.5-0.7 μ m in the bulk sheet. These thin bands, containing extremely fine cells, were present throughout the sheet thickness, at the interfaces between the bonded layers. The bands decreased in width with increasing ARB cycles, but not necessarily in proportion to the strain as they appear harder than the matrix. With wire brushing surface preparation, ARB processing therefore leads to a heterogeneous layered microstructure through the sheet thickness, with the grain structure developed during deformation in the bulk of the sheet being segregated by thin bands (< 3 μ m) of fine lamellar grains and cells delineating previous bond-lines.



Figure 2: Bright field TEM images showing the microstructure in AA1100, at an interface formed in the first roll pass after 7 ARB cycles ($\varepsilon = 9.6$) at a) low magnification and b) high magnification.

The results above do not allow the fine cells observed at the interfaces between bonded layers to be distinguished as grains or subgrains. To alleviate this problem, selected area diffraction (SAD) patterns were collected in the TEM, to analyse the orientation spread present within the wire brushed interface layer. Figure 3 shows SAD patterns, and the corresponding areas they were obtained from, within a typical interface layer and the bulk sheet. It is evident from the bright field image (Figure 3a) that a significant amount of material was missing from the interface and this was due to preferential electropolishing during preparation of the TEM foils. However, the SAD patterns obtained from the areas shown in Figure 3a still reveal that the material at the interface contained a spread of a number of different orientations. Although the spread in orientations at the interface maybe partially caused by a distribution of subgrain orientations, it does indicate that the lamellar structures at the interfaces between layers were probably elongated nano-scale grains.

After several ARB cycles, the processed sheet would therefore, effectively be a composite of bands of nano-scale grains within a matrix containing larger submicron grains [4].

3.3 Comparison between Wire Brushing and Chemical Etching

It has now been established that using wire brushing to prepare sheet surfaces prior to roll-bonding introduces bands of nano-scale grains in the material, situated at the interfaces between bonded layers, resulting in a heterogeneous microstructure through the sheet thickness. If a surface preparation method, such as a chemical etch, was used to remove the oxide instead, then fine grained bands would not be incorporated into the sheet and an ARB processed sheet with a more homogeneous microstructure could potentially be produced. However, the main objective of the surface preparation is to remove the oxide and roughen the surface, enabling a consolidated bond. To examine this aspect more closely, a comparison of the bonding achieved during ARB processing was made between a wire brushing and a hot caustic etch method used for surface preparation. Optical micrographs, showing the central bond-line and surrounding microstructure after 6 ARB cycles at 150°C, using both surface preparation methods are shown in Figure 4. A clear difference in bond quality can be seen between the two surface preparation methods.



Figure TEM 3. images of AA1100 processed to 7 ARB cycles **(ε =** 9.6) showing a) a bright field image of an interface between bonded layers, b) an SAD pattern typical of the bulk material and c) an SAD pattern of wire brushed material at the interface bonded between layers.

At this lower magnification, wire brushing resulted in a central bond line from the last pass that could only just be resolved. Additionally, the bond-lines from previous ARB cycles were very difficult to resolve optically, suggesting they became more consolidated with increasing stain. Contrastingly, using a hot caustic etch to remove the oxide layer and roughen the sheet surface, resulted in poor bonding in places, with large (> 200 μ m) voids appearing between the semi-bonded sheets. These voids would probably decrease the strength of the sheet and provide targets for crack propagation. The voids are also associated with turbulent flow in the matrix and shear banding which can affect the deformation structure of the bulk sheet. The reason behind the poor bonding, achieved when using a hot caustic etch compared to wire brushing, is related to the nature of the prepared surface. It was noted earlier that wire brushing produces an extremely rough, uneven surface containing hills, troughs and overhangs which interlock during roll-bonding

to produce an efficient bond. However, a chemical etch treatment would remove the surface more evenly and produce a more isotropic surface topography, so that the surfaces cannot not interlock as easily during roll-bonding. Additionally, if pitting occurred, the bonding would probably become worse because voids would be created.

3.4 Inclusion of Oxide Debris during ARB Processing

Although surface preparation methods are designed to remove the oxide layer prior to rollbonding, in the case of aluminium the immediate formation of a passive Al_2O_3 layer, means that some oxide will undoubtedly become incorporated and dispersed through the bulk of the sheet during ARB processing. The newly formed oxide layer on pure aluminium is known to be very thin, only ~2.5 nm thick [7], and increases slowly with time. However, the oxide layer can grow during high preheats used during ARB processing. It is currently unclear how much oxide is present in ARB processed sheets. TEM analysis was used to investigate the presence of oxide particles around the interfaces between bonded layers in ARB processed AA1100.



Figure Optical 4: micrographs showing the central bond-line and surrounding microstructure in AA1100 after 6 ARB cycles ($\epsilon = 8.8$) at 150°C when using a) wire brushing and b) a hot caustic etch to prepare sheet surfaces prior to rollbonding.

In addition to the individual reflections in the SAD pattern in Figure 3c, a weak ring pattern can be observed, which is indicative of an amorphous oxide and shows that aluminium oxide, may be present in the wire brushed layer. Further TEM dark field imaging was carried out to ascertain whether oxide particles could be detected at the interfaces. Figure 5 shows a bright field image of another bond interface in the same material and a sequence of dark field images, produced as the transmitted beam was progressively tilted away from the optic axis avoiding strong Al-reflections, so that Bragg condition would intercept any oxide rings. In the sequence of images in Figure 5b-d corresponding to increasing tilt, several small bright particles can be seen, near the interface between the bonded layers. Unfortunately, due to their small size and volume fraction, EDX analysis of the particles was inconclusive and the presence of other contaminates cannot be ruled out. However, it is highly probable that small oxide particles from the wire brushing and roll bonding process are present within the bulk of the sheet.



Figure 5: TEM images of an interface between bonded layers in AA1100 after 7 ARB cycles ($\varepsilon = 9.6$) showing a) a bright field image and b), c), d) a sequence of dark field images produced as the objective aperture was moved out from the central optic axis.

4. Conclusions

Experiments conducted to investigate the effect of surface preparation during ARB processing found that wire brushing resulted in an irregular interface layer of bands nanoscale lamellar grains being incorporated within the sheet centre during roll-bonding. The irregular nature of the layers was caused by folding and flakes formed during the wire brushing process and bonding was not fully complete even after a large number of rolling cycles. After several ARB cycles, and the accumulation of bonded layers, bands 1-3 µm thick of nano-scale grains were distributed throughout the sheet thickness, producing a layered composite structure of the severely deformed original sheets with a coarser ~ 0.7 µm grain/subgrain structure separated by thin nano-grain interface layers. When using a hot caustic etch treatment, instead of wire brushing, to prepare the surfaces for ARB processing, it was found that much poorer bonding was achieved between the sheets and large voids formed at the interfaces between layers. The reduced bonding was attributed to the lower surface roughness and etch pits in the chemically prepared surface. Although TEM investigations were inconclusive as to whether aluminium oxide particles were observed at the interfaces between the bonded layers, the immediate formation of a passive aluminium oxide film when exposed aluminium is subjected suggests oxide will be present within the sheet. It is evident that these microstructural features have important implications for the mechanical and annealing behaviour of ARB processed materials.

References

- R. Z. Valiev, R. K. Islamgaliev and I. V. Alexandrov, Progress in Materials Science, Vol. 45, Iss. 2, [1] pp103-189, 2000. Y. Saito, N. Tsuji, H. Utsonomiya, T. Sakai and R. G. Hong, Scripta Materialia, Vol. 39, Iss. 9, pp1221-
- [2] 1227, 1998.
- N. Tsuji, Y. Saito, H. Utsonomiya and S. Tanigawa, Scripta Materialia, Vol. 40, Iss. 7, pp795-800, [3] 1999.
- [4] C. P. Heason and P. B. Prangnell, International Conference on Textures of Materials (ICOTOM13), Seoul, South Korea, pp733-738, 2002. N. Tsuji, Y. Saito, S-H. Lee and Y. Minamino, Advanced Engineering Materials, Vol. 5, No. 5, pp338-
- [5] 344. 2003.
- G. M. Scamans, A. Asfeth, G. E. Thompson and X. Zhou, 8th International Conference on Aluminium [6] Alloys (ICAA8), Cambridge, UK; pp1461-1466, 2002.
- [7] I. J. Polmear, Light Alloys, Metallurgy of the Light Metals; Third Edition; Arnold, 1995.