# Ultra-fine grained Aluminium Sheets Prepared by Accumulative Roll Bonding

M. Slámová[1,a], P. Homola[1,2,b], M. Karlík[2,c], M. Cieslar[3,d], J. Čížek[3,e]

[1] VÚK Panenské Břežany, s.r.o., Panenské Břežany 50, 250 70 Odolena Voda, Czech Republic
[2] Czech Technical University, Faculty Nucl. Sci. & Phys. Eng., Trojanova 13, 120 00 Praha 2, CZ
[3] Faculty of Mathematics and Physics, Charles University, Ke Karlovu 3, 121 16 Praha 2, CZ
[a]slamova.vuk@volny.cz, [b]vuk@volny.cz, [c]Miroslav.Karlik@fjfi.cvut.cz, [d]cieslar@met.mff.cuni.cz, [e]jcizek@mbox.troja.mff.cuni.cz

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**Abstract.** Accumulative Roll Bonding (ARB) enables the production of large amounts of ultra-fine grained (UFG) sheets and is thus a promising method for industrial applications. The process involves repetitions of surface processing, stacking, rolling, and cutting. The rolling bonds the sheets and after 6 to 8 cycles, UFG materials with high strength and good ductility are produced. Fine second phase dispersion and small grain size are intrinsic features of continuously cast sheets making them good starting materials for ARB. The paper presents the experience with ARB processing of high purity aluminium at room temperature and of hot ARB of continuously cast AlFeMn sheets. In pure Al, hardness increases significantly after two cycles and it raises only a little or decreases during subsequent cycles. Positron lifetime measurements reveal a substantial increase of dislocation density after the first cycle and moderate increase or decrease with further cycles. The high fraction of positrons trapped at grain-boundary dislocations gives evidence for substantial grain refinement confirmed by TEM examinations. Grain size after 4 cycles was estimated by orientation imaging microscopy. The effect of rolling temperature on the quality of roll bonding, grain refinement, and hardness was studied in AlFeMn sheets. After two cycles at 200°C, a mean grain size of 0.4 to 0.8 μm is achieved, but areas with extremely fine grains of 0.1 to 0.3 μm in diameter are also found. Trends of hardness increase similar to these in high purity Al are observed also in AlFeMn alloy. ARB processing of AlFeMn sheet at 250, 300 and 350°C results in better bonding but in smaller increase in hardness due to grain coarsening and partial recrystallization during heating.

## Introduction

Accumulative Roll Bonding (ARB) [1] allows prepare large amounts of bulk ultra-fine grained (UFG) sheets with improved strength and is thus a promising method for industrial applications. ARB involves multiple repetitions of surface processing, stacking, rolling, and cutting. The rolling bonds the sheets and after 6 to 8 cycles, UFG materials with high strength and relatively high ductility are produced. ARB has been successfully used to prepare UFG sheets from different ingot cast aluminium alloys [1]. However, alloys issued from continuous twin-roll casting (TRC) exhibit finer second phase dispersion and smaller grains than ingot cast alloys [2]. They are thus regarded as good starting materials for ARB and are expected to have thermally stable UFG structures. Sheets of thickness 1.0 to 2.2 mm of TRC AlMg3, AlFe1.4Mn, AlFe0.7Si0.7 alloys have been ARB processed at 200°C. The bonding of AlMg3 and AlFe0.7Si0.7 sheets was successful only during the first pass. During the second and third ARB passes, poor bonding or intensive edge cracking occurred in these alloys due to intensive work hardening. High purity aluminium was successfully ARB processed at room temperature (RT). TRC AlFe1.4Mn sheets were successfully AR-bonded up to the 5$^{th}$ cycle at 200°C and up to the 6$^{th}$ cycle at higher temperatures (250, 300, 350°C). The paper presents results of the investigation of ARB processed high purity Al and AlFeMn alloy.

**Experimental**

Twin-roll cast, homogenised and cold rolled sheets of 1.0 or 2.5 mm thickness of the commercial alloy AA8006 (Table 1), and hot rolled 9.0 mm plate of high purity aluminium (Al99.99 - AA1199), were used in the experiments. AA1199 plate was cold rolled to thickness of 2.0 mm. Recrystallized materials were prepared by annealing for 0.5 h at 400°C (AA8006) and 350°C (AA1199). ARB processing consisted in the repetition of 5 steps: 1) surface degreasing in tetrachlorethylene and wire-brushing with 0.3 mm steel wire brush; 2) stacking of two pieces of 300 x 50 x 2.5 mm; 3) joining by Al wires; 4) heating in an electrical furnace to 200, 250, 300 and 350°C (only AA8006); 5) bonding by rolling without lubricant to 50% reduction in thickness. Roll diameter of 340 mm and peripheral speed 0.7 m×min$^{-1}$ were applied in all cases. In order to prevent the propagation of edge cracks, specimen edges were trimmed and smoothed down.

Table 1. Chemical composition of AA8006 alloy [wt.%].

| *Element* | *Mn* | *Fe* | *Si* | *Cu* | *Mg* | *Zn* | *Ti* | *Al* |
|---|---|---|---|---|---|---|---|---|
| *Content* | 0.40 | 1.51 | 0.16 | 0.006 | 0.003 | 0.012 | 0.014 | Balance |

Vickers hardness HV10 measurements on sheet surface were used for evaluating the strength of processed materials. The initial and deformed microstructures were examined using light (LM) and transmission electron microscopy (TEM). LM observations were carried out in the plane normal to the long transverse direction (TD-plane) on samples oxidised in Barker's solution by electrolytic etching. TEM foils 3 mm in diameter were prepared in the rolling plane (ND-plane) by electrolytic twin-jet polishing (-30°C, 30 V) using 6% solution of $HClO_4$ in methanol. The average subgrain diameter $d$ in ARB processed samples was estimated by measurement of the mean intercept length $l_L$ on TEM micrographs (in ND-plane) and calculated according to the relation: $d = K.\ l_L$, $K$ is a constant equal to 1.78 for grains of uniform size of tetrakaidecahedron shape [3]. Positron lifetime (PL) measurements [4] were used to estimate the density and arrangement of dislocation in AA1199 samples. A positron source $^{22}Na$ and a fast-fast PL spectrometer [5] with timing resolution of 160 ps at coincidence count rate 120 s$^{-1}$ were employed. Electron back scatter diffraction (EBSD) and Orientation Imaging Microscopy (OIM) in a scanning electron microscope (SEM) with conventional electron gun was used for grain size estimation in AA1199 sample processed up to 4 cycles. An area of 55 x 35 μm$^2$ was scanned by a step of 0.2 μm in the TD-plane. The grain size was estimated on the registered orientation map using line scans in RD and ND along 5 lines in each direction.

**Results and Discussion**

High purity Al sheets were easily ARB bonded at RT. The hardness of sheets increases from 14.1 to 40.1 kg/mm$^2$, i.e. 2.8 times. Significant hardness increase is observed after the first and second cycles but only a small increase or even a slight softening is observed after subsequent cycles (Fig. 1a).

After initial failures to achieve good roll bonding, five successful ARB cycles were performed at 200°C with AA8006 alloy. The results of those experiments are reported in [6] and are compared with the results of ARB processing at higher temperatures in this paper. ARB at 200°C results in hardness increase from 28 to 58 kg/mm$^2$ (after two cycles), i.e. by a factor of 2.2 (Fig. 1b). HV10 rises a little during subsequent cycles up to 61.4 kg/mm$^2$ (Fig. 1b). Therefore, the relative increase in hardness produced by ARB at T = 200°C in AA8006 specimen is lower as compared to high purity Al, however the absolute hardness achieved is higher. Better roll bonding is obtained when AA8006 sheets are processed at 250, 300 and 350°C, but smaller increase in hardness is achieved (Fig. 1b). At 350°C, HV10 increases from 30 to 49 kg/mm$^2$ during the first cycle. No increase or

softening is observed during subsequent cycles (Fig. 1b). Processing at lower temperatures (250, 300°C) leads to small improvement of ARB strengthening. The maximum hardness is achieved again after 2 cycles, slight softening is observed after subsequent cycles. The maximum relative increase in hardness in AA8006 sheets processed at T ≥ 250°C is not bigger than 1.7 times the initial value. Therefore, ARB induced strengthening is not stable at temperatures above 250°C.

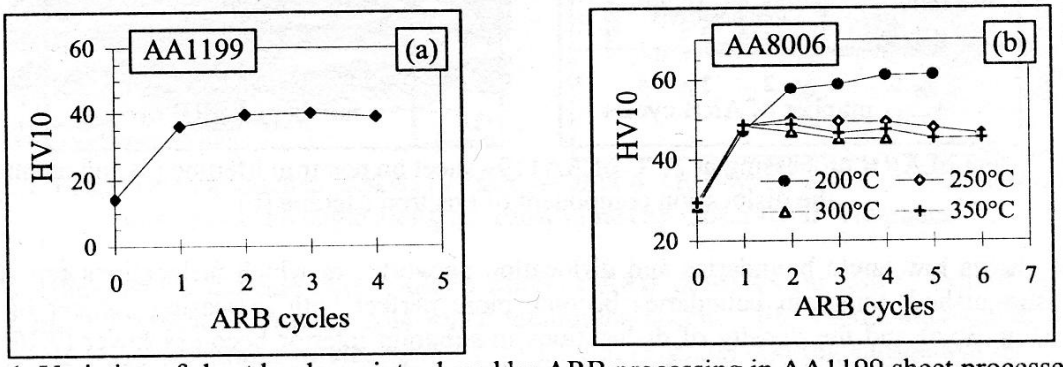


Fig. 1. Variation of sheet hardness introduced by ARB processing in AA1199 sheet processed at 20°C (a) and in AA8006 sheets processed at 200°C to 350°C (b).

LM and TEM examinations indicated that the initial AA1199 sheet was fully recrystallized with grain size of 45 μm (Table 2) and very low dislocation density of $10^{12}$ $m^{-2}$. The input AA8006 sheets used for ARB processing were recrystallized with grain size of less than 20 μm in the rolling direction (RD) and less than 15 μm in ND (Table 2). Thus, the grain size in RD of AA1199 initial material was more than twice larger than in TRC AA8006 sheets.

Table 2. Grain (subgrain) size of initial and ARB processed AA1199 and sheets AA8006 [in μm].

| | $l_L$ (initial, LM) | | $d$ (ARB processed, TEM) | | | | | |
|---|---|---|---|---|---|---|---|---|
| Alloy - T of ARB | RD | ND | 1 cycle | 2 cycles | 3 cycles | 4 cycles | 5 cycles | 6 cycles |
| AA1199 - 20°C | 46 | 38 | 2.0 | 1.4 | 1.2 | 1.7 | - | - |
| AA8006 - 200°C | 15 | 12 | 1.0 | 1.0 | 0.6 | 0.6 | 0.5 | - |
| AA8006 - 250°C | 15 | 12 | 1.2 | - | 1.0 | - | - | 1.2 |
| AA8006 - 350°C | 19 | 14 | 1.3 | - | 1.3 | 1.3 | 1.3 | 1.3 |

ARB processed AA1199 samples were first studied by PL measurements. Their PL spectra could be fitted by two exponential components with lifetimes $\tau_1$ and $\tau_2$. The shorter component $\tau_1$ is the contribution of free positrons, while $\tau_2$ is a contribution of positrons trapped at defects. Figure 2a shows the evolution of $\tau_1$ and $\tau_2$ with increasing ARB cycles. It can be seen that $\tau_1$ decreases as result of ARB processing, whereas $\tau_2$ is almost unchanged. According to previous measurements in pure Al [7], it can be assumed that $\tau_2 \approx 243$ ps is the lifetime of positrons trapped at dislocations introduced by ARB processing. Figure 2b shows that the relative intensity $I_2$ of the dislocation component drastically increases after the first ARB cycle but changes only moderately during subsequent ARB cycles. This is indicative for a substantial increase of dislocation density during the first ARB cycle, while further cycles do not cause any significant change. The application of the two-state trapping model [4] to the data reveals that the dislocations in ARB processed samples are distributed non-homogeneously, i.e. they are probably arranged in cell and subgrain boundaries. In accordance with PL measurements and modelling, TEM examinations after the 1. cycle show well-defined cells and subgrains of size 1-2 μm (Fig. 3a) and low dislocation density in their interiors ($3.10^{13}m^{-2}$). However, subgrain formation due to dynamic dislocation recovery is not complete.

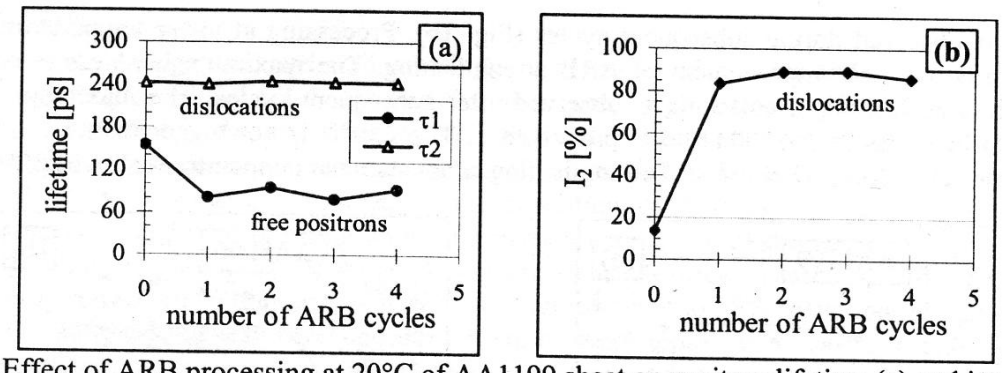


Fig. 2. Effect of ARB processing at 20°C of AA1199 sheet on positron lifetime (a) and intensity of the dislocation component of positron lifetime (b).

Fig. 3a shows low angle boundaries and dislocation networks, in which dislocations can still be well distinguished. Subgrain boundaries become more perfect with increasing number of ARB cycles (Fig. 3b,c) and the density of dislocations in subgrain interior becomes lower ($7.10^{12}m^{-2}$). After the third and fourth cycles, small recrystallization nuclei are also observed. The subgrain size after 3 cycles is of 1.2 μm and it even increases to 1.7 μm during the next cycle (Fig. 3c, Table 2). Thus, the subgrains in AA1199 ARB sheets processed at RT are much larger and deformation recovery more pronounced than in AA8006 sheets processed at 200°C (see below) probably due to its large initial grains and absence of particles, which could pin grain boundaries.

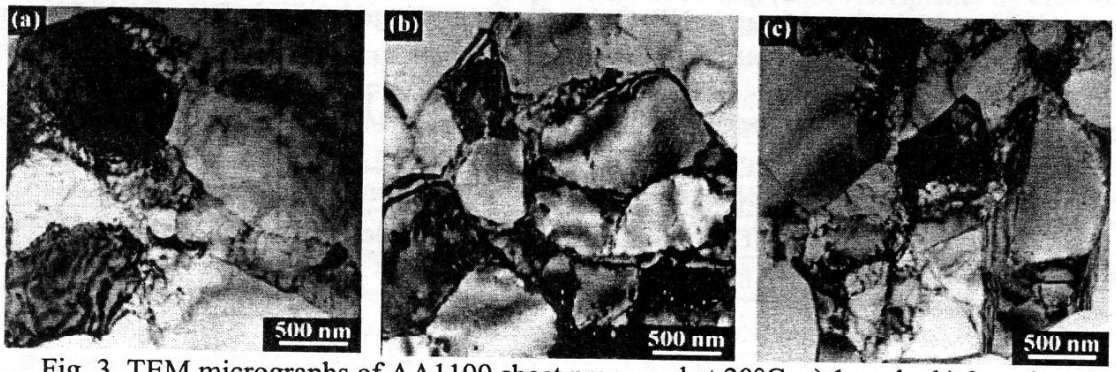


Fig. 3. TEM micrographs of AA1199 sheet processed at 20°C: a) 1 cycle; b) 3 cycles; c) 4 cycles. Observed in ND-plane.

EBSD measurements were carried out only on the sample ARB processed to true strain $\varepsilon = 3.2$ (4 ARB cycles). Fig. 4a shows the orientation map and Fig. 4b grain misorientation found along one of the 5 scanned lines in RD, respectively. The automatically calculated grain size is 0.95 μm. However, the automatic procedure probably strongly underestimates the actual grain size. One reason for this discrepancy may be that in a deformed sample there are many positions with low confidence index and image quality coefficient. Moreover, the automatic procedure does not take into account the anisotropic shape of the grains. The semi-automatic mean intercept length procedure yields an average grain size of 5.5 μm and 1.9 μm in RD and ND, respectively. Only boundaries with misorientation > 15° were considered. The latter values are much more probable than the automatically calculated one. The subgrain size estimated from OIM is 2.5 μm and 1.1 μm in RD and ND, respectively. The former corresponds to the grain diameter *d* estimated from TEM micrographs (Table 2). However, the value 2.5 μm is not in good agreement with TEM estimation. The most probable reason of this discrepancy is the insufficient OIM spatial resolution of conventional SEM when UFG materials are measured. Better results should be obtained with a field emission gun (FEG) SEM.

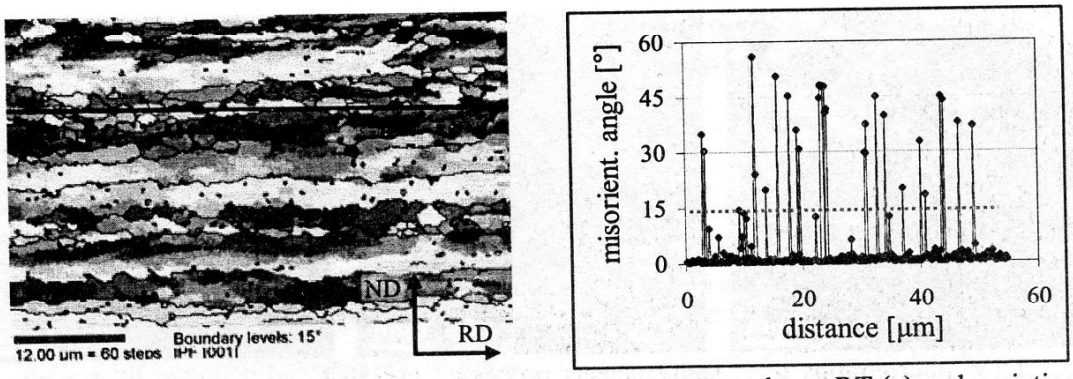


Fig. 4. EBSD orientation map of AA1199 sample after 4 ARB cycles at RT (a) and variation of misorientation angle along the shown line (b). Observed in TD-plane.

LM examinations indicate that AA8006 samples ARB processed at 200°C exhibit deformed grain structure typical for heavily cold rolled aluminium sheets. TEM examinations after the first ARB cycle reveal subgrains of size from 0.5 to 1.5 µm (Fig. 5a). During subsequent cycles, low-angle boundaries convert to high-angle boundaries (Figs. 5b,c) appearing in a typical fringe contrast (Fig. 5b). The size of the majority of grains is in the range from 0.4 to 0.8 µm, the largest grains are of 1.2 µm in diameter. Dislocation density in subgrains or grains remains almost unchanged throughout all cycles of ARB processing and is indicative for a recovered substructure. In the samples with 2$^{nd}$ and 5$^{th}$ cycles, 150 µm wide areas with much finer grains (0.1 to 0.3 µm in diameter) are observed (Fig. 5b). X-ray energy dispersion analysis does not show any difference in matrix composition in these areas, neither fine-dispersed particles that could pin grain boundaries are observed. Tsuji et al. [8] suggest that these extremely refined grains are formed as result of the intensive friction and shear deformation involved in surface brushing.

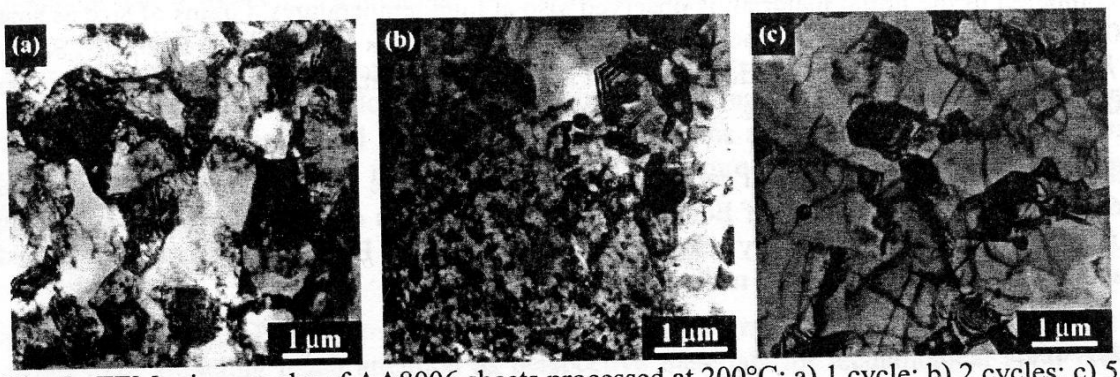


Fig. 5. TEM micrographs of AA8006 sheets processed at 200°C: a) 1 cycle; b) 2 cycles; c) 5 cycles. Observed in ND-plane.

In contrast to AA8006 specimens ARB processed at 200°C, coarser grains form in specimens processed at 350°C. Post-processing examinations by light microscopy (LM) show grains elongated in RD, but after the 2$^{nd}$ and further cycles the grain size in ND is much larger than that expected after several ARB cycles, which would introduce significant plastic deformation. TEM examinations (Table 2, Fig. 6) confirm that the average grain size refines in the 1$^{st}$ ARB cycle down to 1.3 µm and it remains almost unchanged by further ARB processing. However, areas of grain size as small as 500 nm are also locally observed. Both LM and TEM indicate that subgrain coarsening and recrystallization occur during the heating to 350°C. These processes cause the low sheet hardening achieved by the ARB processing at 350°C. The situation is similar also during processing at 250°C and 300°C, where deformation recovery is also observed and is more pronounced at larger strains.

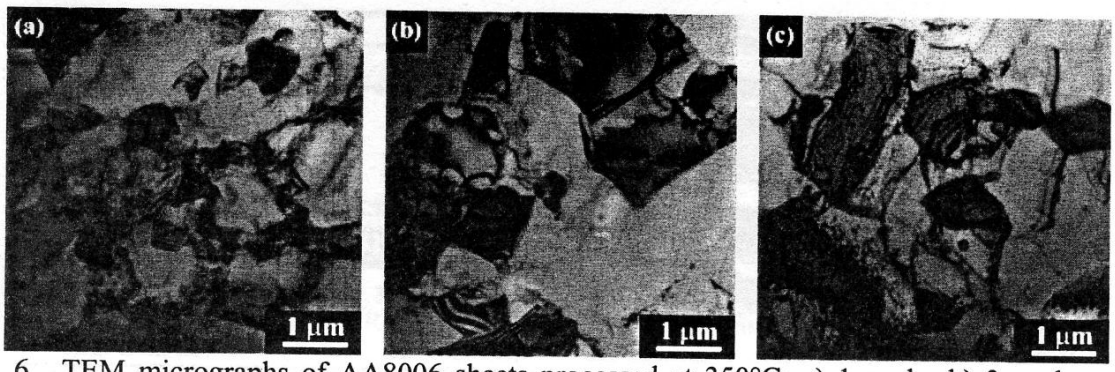


Fig. 6. TEM micrographs of AA8006 sheets processed at 350°C: a) 1 cycle; b) 3 cycles; c) 5 cycles.

## Conclusions

Accumulative roll bonding at temperatures between 200°C and 350°C was applied in order to prepare ultra-fine grained materials from continuously cast AA8006 aluminium alloy and at 20°C from direct chill cast high purity aluminium (AA1199), respectively. ARB processing of AA1199 introduces more than twofold increase in hardness due to refinement of grain size from 45 to 1 μm after 3 cycles. PL measurements show increase in dislocation density only during the first two cycles. PL results are in good agreement with TEM observations. EBSD is a powerful technique for grain size estimation of UFG materials. However, adequate space resolution achieved by FEG SEM is indispensable. Roll bonding of AA8006 sheets is more successful at 250, 300 and 350°C than at 200°C. However, ARB at higher temperatures does not result in as significant grain refinement and strength increase as does processing at 200°C. Subgrain coarsening and recrystallization occurring during heating are the probable causes of this unsuccess. Nevertheless, significant grain refinement, as compared to the initial material, is observed also at high temperatures. Grains of size as small as 100 nm are produced in part of the material subjected to 5 cycles of processing at 200°C. Six cycles of ARB processing at 350°C produce grains of mean size 1.3 μm, but areas of grain size as small as 500 nm are also locally observed.

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